



Coherent optical control of correlation waves of spins in semiconductors

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We calculate the dynamical fluctuation spectrum of electronic spins in a semiconductor under a steady-state illumination by light containing polarization squeezing correlations. Taking into account quasiparticle lifetime and spin relaxation for this nonequilibrium situation we consider up to fourth order optical effects which are sensitive to the squeezing phases. We demonstrate the possibility to control the spin fluctuations by optically modulating these phases as a function of frequency, leading to a non-Lorentzian spectrum which is very different from the thermal equilibrium fluctuations in n -doped semiconductors. Specifically, in the time-domain spin-spin correlation can exhibit time delays and sign flips originating from the phase modulations and correlations of polarizations, respectively. For higher light intensity we expect a regime where the squeezing correlations will dominate the spectrum.

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I. INTRODUCTION

When a semiconductor absorbs circularly polarized light an average collective spin polarization $\langle \mathbf{S}(t) \rangle$ is induced in the conduction band—a phenomenon known as *optical orientation*.^{1,2} In direct-gap III-V semiconductors this is a result of optical transitions of electric dipole type across the electron-hole gap. In this process there is a net transfer of angular momentum from the helicity of the photons to the angular momentum of the electrons. The optical selection rules are such that for the circularly polarized light the excitation rate for electrons with one spin projection is larger compared to the other, resulting in a net spin polarization in the conduction band. Following the post excitation spin dynamics one can investigate spin-relaxation mechanisms using techniques such as time-resolved Faraday rotation and time-resolved photoluminescence.^{3,4} It is also possible to monitor the space resolved distribution of spins, their orientation and magnitude, as well as coherently control these quantities.^{5–7}

The next interesting object to study is the fluctuation of the spin in a semiconductor. In thermal equilibrium these fluctuations were measured by employing the Faraday effect.⁸ A linearly polarized probe beam that passes through the sample is affected by the instantaneous magnetization of the sample, and its vector of polarization acquires a rotation, proportional to the magnetization.⁹ By measuring the spectrum of the polarization fluctuations, it is possible to relate it to the spin fluctuations.

Here we propose to go beyond the equilibrium and to study dynamical fluctuations of the electronic spins which are induced by external optical field. In the way similar to the average spin induced by the polarization of absorbed light, we wish to consider how fluctuations of the light polarization produce upon absorption dynamical fluctuations of electronic spins. We will consider polarization-squeezed light which has a predetermined spectrum of two-photon correlations. We find that the dependence of the spin structure factor $\langle \langle S(t)S(t') \rangle \rangle$ on the time difference $t-t'$ is sensitive to the phase of the squeezing correlations of the light [Eq. (29)]. We demonstrate that by modulating the frequency depen-

dence of the phases, the dependence on $t-t'$ can deviate significantly from exponential behavior (Figs. 5 and 6). Since in practice these phases indeed can be modulated, this opens new possibilities for a coherent control of spin fluctuations in bulk semiconductors.

In the past theoretical suggestions to observe quantum optical effects in light-matter interaction involved models with squeezed radiation reservoirs interacting with atoms and semiconductors.^{10–12} This, however, requires a high quality squeezed reservoir, which experimentally remains challenging to achieve. Another quantum optical effect which does not involve a reservoir is related to two-photon absorption of squeezed light by a three-level system^{13,14} and was demonstrated experimentally by Georgiades *et al.*¹⁵ Other works explored the aspect of transmission of squeezed light through bulk media^{16–18} and photoionization.¹⁹ Schemes for transferring correlations from light to matter have recently been developed in atomic and molecular optics (AMO) both theoretically^{20,21} and experimentally.^{22,23} These schemes employ either coherent optical dipoles of atomic V systems or ground-state coherence of Λ systems, leading to a *second-order* dependence of the spin fluctuations on the squeezed optical field.

Unfortunately, in semiconductors one is faced with strong dephasing of optical dipoles as well as valence-band spins due to Coulomb, electron-phonon, and spin-orbit interactions, rendering the above atomic optics schemes impractical. We note, however, that the dephasing and relaxation of the conduction-band spins are much slower. In addition, continuous energy bands in semiconductors have very different level structure and optical selection rules. In contrast to the AMO schemes, we suggest using this and employing *fourth order* optical effects to manipulate the collective spin of the conduction-band electrons through the process of two-photon absorption.

For simplicity and to isolate the transfer of fluctuations from the average, we assume in this work that the semiconductor is irradiated with the light which is on the average unpolarized but possesses nonvanishing squeezing correlations between different polarization amplitudes. Such light (polarization-squeezed vacuum) was discussed by

Karrasiov,²⁴ Lehner *et al.*,²⁵ and Korolkova *et al.*²⁶ and generated in several experiments.^{27,28} In this paper we demonstrate [see Eq. (29)] that by externally manipulating the phase of the frequency dependent photonic correlations a possibility opens up of optical coherent control of spins in semiconductors. The essence of this effect lies in controlling the interference between quantum amplitudes related to many pairs of different optical frequencies. We predict that in order to observe this effect it is not necessary to have squeezing in quantum sense, i.e., below the shot-noise limit. This should allow for easier measurements since such light can be generated with higher intensities. Nevertheless we anticipate that also in the quantum optical regime interesting features should appear in the spectrum following our previous predictions for the static spin correlations.²⁹

The feasibility of observing spin effects in semiconductors relies on the relatively weak coupling of the conduction-band spins to the environment, i.e., slow spin flipping processes. This has been experimentally demonstrated in various situations in the past.^{3,4,30-33} We also rely on the fact that for the holes the situation is different with the corresponding rates estimated to be several orders of magnitude higher.¹⁻³ Accordingly we have neglected the contribution of the hole spins to the total spin correlations. To observe induced spin fluctuations it is necessary to extend the experimental capabilities of measuring spin fluctuations to the nonequilibrium regime. In principle two beams should be employed in such an experiment, i.e., a correlated light pump and a linearly polarized probe acting at the same time. In addition to the measurement of the induced magnetic moment, it is necessary to have phase manipulation capabilities in the incoming pump beam, similarly to those demonstrated recently for squeezed vacuum.³⁴

In this work we have taken into account scattering effects of nonradiative processes in a semiphenomenological way via relaxation times. However, our results do not depend in an essential way on the details of the interactions and the main qualitative features that we demonstrate should be reproducible even in a more detailed study. This claim is supported by a separate diagrammatic calculation³⁵ which reproduces qualitatively the main results that are presented here.

The structure of the paper is as follows. In Sec. II we introduce the two-band model of electrons interacting with the driving field. In Secs. III and IV we calculate the second and fourth order contributions (in the optical field) to the spin fluctuations, assuming zero lattice temperature and phenomenological description of relaxations. In Sec. V we develop physically motivated simplifications of our results and discuss their meaning. In Sec. VI we explore the consequences of phase modulations of the squeezing correlations on the spin-fluctuation spectrum. In Appendix A we give details concerning the dipole matrix elements which appear in the light-matter Hamiltonian. In Appendix B we explain the physical nature of our phenomenological approach and how it affects the results. Finally in Appendix C we discuss the calculation details of additional fourth order contributions which are small and not included in the main calculation.

II. MODEL

Valence and conduction bands of a direct-gap semiconductor (III-V) interacting with the light are modeled by the Hamiltonian

$$H = \sum_{k\sigma} \epsilon_k^c c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k'\sigma} \epsilon_{k'}^v v_{k'\sigma}^\dagger v_{k'\sigma} + H_{LM} + H_C + H_{SO}, \quad (1)$$

where the operators $c_{k\sigma}$ and $v_{k'\sigma}$ denote annihilation operators of the electrons in the conduction and valence bands, with quasimomenta k, k' and total angular momentum σ . The index σ in the second term enumerates both the degenerate valence bands and their spin degeneracy. The effect of the spin-orbit (SO) interaction in the valence bands is usually accounted for by Luttinger model for degenerate bands.³⁶ This model takes into account the mixing of the angular momentum $j=3/2$ states due to spin-orbit coupling. As we explain in Appendix A, we will take into account the heavy-hole valence-band transitions with the energies ϵ_k^v being degenerate with respect to the spin projection and renormalized due to the SO interaction in the valence band. Generally important also are the Coulomb interaction (H_C) between electrons, which is responsible for excitonic and scattering effects, and the spin-orbit interaction in the conduction band (H_{SO}). We will focus here on effects related to the light-matter interaction (H_{LM}) in the dipole approximation. We study optically excited electrons in the conduction band with energies above the ionization level of the exciton. These electrons are well described as quasiparticles with a finite lifetime arising from the momentum scattering induced by disorder and Coulomb interactions. We will show in Appendix A that to a good approximation it is possible to write the light-matter interaction as

$$H_{LM}(t) = \sum_{k,p,\sigma} [d_{k,p}^\sigma P_{k,k+p,\sigma}^\dagger b_{p,\sigma} e^{i\Omega_{k,p}t} + d_{k,p}^{\sigma*} b_{p,\sigma}^* P_{k,k+p,\sigma} e^{-i\Omega_{k,p}t}], \quad (2)$$

where the operator $P_{k,k+p,\sigma} = v_{k,\sigma}^\dagger c_{k+p,\sigma}$ is the interband polarization. $\Omega_{k,p} = \epsilon_{k+p}^c - \epsilon_k^v - \omega_p$ are the differences between quasiparticle energies and the photons and $b_{q,\sigma}$ are optical-field amplitudes with wave number q and polarization σ . For these transitions it is sufficient to use one index (σ), denoting projection of conduction-band spin ($\pm 1/2$), projection of valence-band total angular momentum ($\pm 3/2$), and helicity (± 1), depending on the context where it appears. We also show in Appendix A that for most purposes the interaction transition matrix element $d_{k,p}^s$ can be taken as independent of the direction of k and of s and simply denoted as a constant d .

The spin-orbit Hamiltonian (H_{SO}) is responsible for the D'yakonov-Perel' (DP) and Bir-Aronov-Pikus (BAP) spin-relaxation mechanisms, which are described by the following effective Hamiltonians.³

$$H_{SO}^{(DP)} = \frac{1}{2} \hbar \sum_{s,s'} [\vec{R}(k) \cdot \vec{\sigma}]_{s,s'} c_{ks}^\dagger c_{ks'},$$

$$H_C^{(\text{BAP})} = A \sum_{k,k',q,s,s',j,j'} (\vec{J} \cdot \vec{\sigma})_{s,s',j,j'} c_{k,s}^\dagger c_{k+q,s}^\dagger v_{k',j}^\dagger v_{k'-q,j'}, \quad (3)$$

where $\vec{R}(k)$ and A are parameters depending on the material, dimensionality, temperature, and details of the optical excitation. We discuss in Sec. III the main effects of H_{SO} and H_C on the optically excited spin correlations. Note that the Coulomb interaction between electrons in the conduction band ($c^\dagger c c^\dagger c$), which is not included, does not affect the relaxation of the total spin, since it is not interacting with external degrees of freedom. Even though in principle there are additional spin-relaxation mechanisms such as Elliot-Yafet, the dominant mechanisms for spin relaxation are D'yakonov-Perel' and Bir-Pikus³ when the electron gas is nondegenerate (nonmetallic regime). For observing the effects which we discuss here, materials with a relatively long spin lifetime such as GaAs are advantageous. Spin-relaxation times of the average spin have been measured by different techniques for different materials and experimental conditions.^{3,30} These include n -GaAs quantum wells with different widths,⁴ doping levels,³⁷ materials,^{31,32} as well as bulk n -GaAs.³³ In these experiments spin lifetimes ranging from hundreds of picoseconds to tens of nanoseconds have been reported. In some cases the long lifetimes were measured even in room temperature.³⁰ The contribution of the valence-band holes to the driven spin fluctuations is assumed throughout to be small. This assumption is realistic in bulk semiconductors from the group III-V, where the spin-flip rates for the holes are up to 3 orders of magnitude faster than for the conduction-band electrons,³ with estimates and measurements placing the hole spin lifetime at around 1 ps. These rates lead to very broad distributions of the holes spin fluctuations in Fourier space with negligible contributions near $\omega=0$. It is very difficult in experiments to capture such (10^3 GHz) fast oscillations, so in a realistic experiment we can neglect their effect.³⁷

Polarization properties of photons are described by the Stokes parameters^{26,38} which in the circular polarization basis are written as time averages of

$$p_i = \sum_{q,q',\lambda,\lambda'} b_{q\lambda}^* (\sigma_i)_{\lambda\lambda'} b_{q'\lambda'}, \quad (4)$$

where $\sigma_{i=0,\dots,3}$ denote the unit and Pauli-spin matrix. The averaging is over a time which is longer than the typical correlation time of the field. We consider a collinear pump beam with a range of frequencies $\omega_0 \pm B/2$ above the electron-hole gap and time and bandwidth averaged Stokes parameters $\langle p_0 \rangle = (2\pi c/LB) \sum_{q\lambda} N_{q\lambda}$, $\langle p_1 \rangle = \langle p_2 \rangle = \langle p_3 \rangle = 0$, where $N_{q\lambda}$ is the average photon occupation per mode and L is the mode quantization length. This is unpolarized light with the Stokes vector fluctuating around the origin of the Poincaré sphere. The fluctuations are described by the covariance matrix $p_{ij} = \langle p_i p_j \rangle$, which for a Gaussian-type field depends on the normal $\langle b_{q\lambda}^* b_{q'\lambda'} \rangle$ as well as anomalous $\langle b_{q\lambda} b_{q'\lambda'} \rangle$ correlations; the latter constituting the main characteristics of squeezed light.³⁹ In addition to normal correlations $\langle b_{q\lambda}^* b_{q'\lambda'} \rangle = N_q \delta_{\lambda\lambda'} \delta_{qq'}$ they possess four generic anomalous correla-

tions: two for the same-polarization squeezing $\langle b_{q\pm} b_{q'\pm} \rangle = M_{q\pm}^{(1)} \delta_{q+q',2q_0} \delta_{\omega_q+\omega_{q'},2\omega_0}$ and two for the opposite-polarization squeezing $\langle b_{q\pm} b_{q'\mp} \rangle = M_{q\pm}^{(2)} \delta_{q+q',2q_0} \delta_{\omega_q+\omega_{q'},2\omega_0}$, where $M_{q\pm}^{(1,2)}$ are complex functions and $\omega_0 = cq_0$. It can be shown^{40,41} that for quantum fields $|M_{q\lambda}^{(1,2)}| \leq N_{q\lambda} \sqrt{N_{q\lambda} + 1}$ while $|M_{q\lambda}^{(1,2)}| \leq N_{q\lambda}$ for classical fields, which we discuss here. We will later remark on the possible effects of the quantum regime in Sec. VI. This type of light is also called polarization-squeezed vacuum²⁶ or two-mode squeezed-vacuum state of type II.^{25,42}

We begin by deriving the Heisenberg equations of motion for the spin waves in the conduction band. The spin-density operator⁴³ is given by

$$\begin{aligned} \vec{S}(r,t) &= \sum_{s,s'} \psi_s^\dagger(r,t) \vec{\sigma}_{s,s'} \psi_{s'}(r,t) \\ &= \sum_{s,s'} \sum_{k,k'} e^{-i(k-k')r} c_{k,s}^\dagger(t) \vec{\sigma}_{s,s'} c_{k',s'}(t) \end{aligned} \quad (5)$$

and we define the q component of the spin as

$$\vec{S}(q,t) = \int_V d^3r e^{-iqr} \vec{S}(r,t) = \sum_{k,s,s'} c_{k-q,s}^\dagger(t) \vec{\sigma}_{s,s'} c_{k,s'}(t). \quad (6)$$

Without the optical excitation, the spin correlations are zero when the lattice temperature is $T=0$ because there are no electrons in the conduction band. When the electric field of the light is stochastic with a zero average, so is the average spin component $\langle \vec{S}(q,t) \rangle$ and therefore we study the correlations. As we will discuss in Sec. IV, due to the optical selection rules only the averages $\langle S_z(q,t) S_z(q',t') \rangle$, with $S_z(q,t) = \sum_k [c_{k-q,\uparrow}^\dagger c_{k,\uparrow} - c_{k-q,\downarrow}^\dagger c_{k,\downarrow}]$, are affected by the squeezing correlations of the light beam (directed along the \hat{z} direction) and therefore we will focus on them. The function $\langle S_z(q,t) S_z(q',t') \rangle$ is related to experiment in a similar way that the dipole fluctuations $\langle \sigma(t)_+ \sigma(t')_- \rangle$ of an atom are related to the measurable fluorescence spectrum⁴⁴ through the first-order correlation function of the optical fields,⁴⁵ assuming we have a suitable spectrometer at hand. In our case of the spins one way that comes to mind is making use of the Faraday effect, which is customarily used to measure optically injected spin in semiconductors.

Using the Hamiltonian (1) and (2) the equations of motion which define the spin waves are given by

$$\begin{aligned} \frac{d}{dt} n_{k,k',\sigma}^c &= i \delta \epsilon_{k,k'}^c c_{k,\sigma}^\dagger c_{k',\sigma} + i \sum_p [-d P_{k'-p,k,\sigma}^\dagger b_{p\sigma} \\ &\quad + d^* b_{p\sigma}^* P_{k-p,k',\sigma}] + i [H_{\text{SO}}, n_{k,k',\sigma}^c] + i [H_C, n_{k,k',\sigma}^c], \\ \frac{d}{dt} n_{k,k',\sigma}^v &= i \delta \epsilon_{k,k'}^v v_{k,\sigma}^\dagger v_{k',\sigma} + i \sum_p [d P_{k',k+p,\sigma}^\dagger b_{p\sigma} - d^* b_{p\sigma}^* P_{k,k'+p}] \\ &\quad + i [H_{\text{SO}}, n_{k,k',\sigma}^v] + i [H_C, n_{k,k',\sigma}^v], \end{aligned}$$

$$\begin{aligned} \frac{d}{dt} P_{k',k,\sigma}^\dagger &= \frac{d}{dt} c_{k,\sigma}^\dagger v_{k',\sigma} \\ &= i\Delta \epsilon_{k,k'} c_{k,\sigma}^\dagger v_{k',\sigma} - i d^* \sum_p b_{p\sigma}^* [n_{k,k'+p,\sigma}^c - n_{k-p,k',\sigma}^v] \\ &\quad + i[H_{SO}, P_{k',k,\sigma}^\dagger] + i[H_C, P_{k',k,\sigma}^\dagger], \end{aligned} \quad (7)$$

with $\delta \epsilon_{k,k'}^c = \epsilon_k^c - \epsilon_{k'}^c$, $\Delta \epsilon_{k,k'} = \epsilon_k^c - \epsilon_{k'}^v$, and $n_{k,k',\sigma}^c = c_{k,\sigma}^\dagger c_{k',\sigma}$.⁴⁶

We treat the interaction between the electrons and the optical field in perturbation theory. The spin operator in Heisenberg picture can formally be written as

$$\begin{aligned} \langle S_z(q,t) S_z(q',t') \rangle &= \langle S_z^{(0)}(q,t) S_z^{(0)}(q',t') \rangle + \langle S_z^{(1)}(q,t) S_z^{(1)}(q',t') \rangle + \langle S_z^{(0)}(q,t) S_z^{(4)}(q',t') \rangle + \langle S_z^{(4)}(q,t) S_z^{(0)}(q',t') \rangle \\ &\quad + \langle S_z^{(1)}(q,t) S_z^{(3)}(q',t') \rangle + \langle S_z^{(3)}(q,t) S_z^{(1)}(q',t') \rangle + \langle S_z^{(2)}(q,t) S_z^{(2)}(q',t') \rangle. \end{aligned} \quad (9)$$

However, at $T=0$ the combinations 0×0 , 4×0 , 0×4 are zero since the spin operator (on the “0” side) would be acting on the empty conduction-band state. Therefore we are left with

$$\begin{aligned} \langle S_z(q,t) S_z(q',t') \rangle &= \langle S_z^{(1)}(q,t) S_z^{(1)}(q',t') \rangle \\ &\quad + \langle S_z^{(2)}(q,t) S_z^{(2)}(q',t') \rangle \\ &\quad + \langle S_z^{(1)}(q,t) S_z^{(3)}(q',t') \rangle \\ &\quad + \langle S_z^{(3)}(q,t) S_z^{(1)}(q',t') \rangle, \end{aligned} \quad (10)$$

i.e., only second and fourth order contributions. The former are not affected by squeezing and contribute to the background of the fluctuation spectrum. The latter, however, is affected by the q -dependent phase modulations of the function $M_{q\pm}^{(1,2)}$, through a microscopic process of double absorption (see Sec. IV). This process is illustrated in Fig. 1 showing two cases of opposite and same polarizations. Two quantum amplitudes related to absorption of two different pairs of photons proportional to $M^{(1,2)}(\omega)$ and $M^{(1,2)}(\omega+\Omega)$

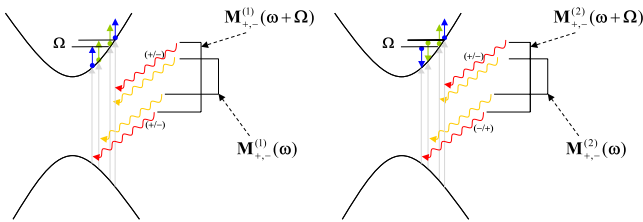


FIG. 1. (Color online) Illustration of double-photon excitation processes (long gray arrows), with yellow and red colors for the two pairs, which excites spin-polarized electrons (short arrows). The left (right) figure describes absorption amplitudes of photons of same (opposite) helicity. The interference of these amplitudes creates a spin-correlation wave with a frequency Ω .

$$\begin{aligned} S_z(q,t) &= S_z(q,t)^{(0)} + S_z(q,t)^{(1)} + S_z(q,t)^{(2)} + S_z(q,t)^{(3)} \\ &\quad + S_z(q,t)^{(4)} + \dots \end{aligned} \quad (8)$$

We shall see in Sec. III that to second order the spin fluctuations are proportional only to the photon occupation $\langle b^*b \rangle$ and therefore are not affected by squeezing, where the latter only makes an effect in the fourth order. When using it to expand $\langle S_z(q,t) S_z(q',t') \rangle$ we have to take all the possible combinations which contain the optical interaction an even number of times, since the spin operators conserve the number of particles

will be shown to appear in interference terms such as $M^*(\omega)M(\omega+\Omega)$ which bring into effect their externally controlled phase difference.

We shall derive the Heisenberg equations of motion up to fourth order in the optical field by writing formal solutions and substituting back to the previous order. Then we will use the resulting expansion to calculate the averages in Eq. (10) order by order, taking into account the nature of the optical field. An important part of the model is the treatment of electron and spin lifetimes, which we will discuss in Sec. III.

III. CALCULATION OF THE SECOND-ORDER CONTRIBUTION

This physical optical process, of the lowest order, is responsible for generating carrier and spin densities in the conduction band of a semiconductor. It is convenient to analyze their space-time profile in Fourier space (q, ω) , given by the correlation function of $S_z(q, \omega)$ operators. The perturbative correction which we discuss here turns out not to depend on squeezing or coherent properties of the light field and will ultimately just serve as a background for the more interesting higher order processes (2×2). At the last stage of the calculation [see Eq. (19)] we neglect the momenta of the spin wave (q) and the photon (p) compared with the typical electronic momentum (k) in places where they appear together as a sum (e.g., $k+p \rightarrow k$). Within this approximation the only angular dependence that remains is that of the dipole matrix element (see Appendix A), which amounts to renormalization of the value of the matrix element. Now we use Eq. (7) to construct the equation for the spin, with $b_{q,\uparrow}$ ($b_{q,\downarrow}$) denoting right (left) polarizations, respectively. The first equation then reads

$$\begin{aligned}
\frac{d}{dt}S_z(q,t) &= \sum_k i\delta\epsilon_{k-q,k}e^{i\delta\epsilon_{k-q,k}t}(n_{k-q,k,\uparrow}^c - n_{k-q,k,\downarrow}^c) \\
&+ \sum_k \left[i \sum_p \left(-d P_{k-p,k-q,\uparrow}^\dagger b_{q\uparrow} + d^* b_{p\uparrow}^* P_{k-p-q,k,\uparrow} \right) \right. \\
&\left. - i \sum_p \left(-d P_{k-p,k-q,\downarrow}^\dagger b_{p\downarrow} + d^* b_{p\downarrow}^* P_{k-p-q,k,\downarrow} \right) \right] \\
&+ i[H_{SO}, S_z(q,t)] + i[H_C, S_z(q,t)]. \quad (11)
\end{aligned}$$

The first term on the right-hand side describes a free evolution of the spin wave with typical frequencies of v_0q , where v_0 is the electron velocity at k_0 , the latter being the quasimomentum of the electron optically excited by the photon of the middle frequency ω_0 . Since the light induces spin correlations with the dispersion of cq , we are physically motivated to neglect the slow free evolution (v_0q) in the following treatment.

Instead of developing a microscopic theory for the relaxation of the spin wave S_z and interband polarization P , we replace the combined effect of H_{SO} and H_C by a quasiparticle description with a width $\gamma = \tau^{-1}$ and a phenomenological re-

laxation $\gamma_s = \tau_s^{-1}$. Under broadband excitation conditions, and when the electrons kinetic energy is large compared to the electron-hole exchange energy, it is reasonable to model them as quasiparticles with a finite lifetime (τ). This lifetime enters the model through the *level width* of the electronic energies ϵ_k^c . In addition to τ it is physically plausible, and supported by many experiments, to assume the existence of another *macroscopic* relaxation time (τ_s) of the spin wave which is usually much longer than the electron lifetime $\tau_s \gg \tau$ for semiconductors such as GaAs. The spin lifetime can be entered as a phenomenological decay term ($-\gamma_s S_z$) in the equation of motion for the spin operator S_z [Eq. (11)], similarly to a decay term in a quantum-Langevin equation of motion. Equivalently, it can enter as an additional imaginary part (broadening) to the frequency ω_q of the spin wave. We explain in Appendix B the limitations of the phenomenological approach and how to reconcile it with the results that we obtained for the static correlations in the previous work.²⁹ The averaging of $\langle S_z(q,t)S_z(q',t') \rangle$ over the ground state of with $T=0$ leaves only the terms $\langle PP^\dagger \rangle$. The general expression reads

$$\begin{aligned}
\langle S_z(q,t)S_z(q',t') \rangle^{(2)} &= |d|^2 e^{-\gamma_s(t+t')} \int_{t_0}^t dt_1 e^{\gamma_s t_1} \int_{t_0}^{t'} dt_2 e^{\gamma_s t_2} \\
&\times \sum_{k,k'} \sum_{p,p',\sigma,\sigma'} (-1)^{\sigma-\sigma'} e^{i(\omega_{p't_1} - \omega_{p't_2})} \langle b_{p\sigma}^* b_{p'\sigma'} \rangle \langle P_{k-p-q,k,\sigma}(t_1) P_{k'-p',k'-q',\sigma'}^\dagger(t_2) \rangle, \quad (12)
\end{aligned}$$

where t_0 is the initial time when the system was in the ground state before the optical fields were turned on. Given that $N_{p\sigma}$ is the photon occupation function, γ is the quasiparticle lifetime and $\sigma, \sigma' = \pm 1/2$; we also have

$$\langle b_{p\sigma}^* b_{p'\sigma'} \rangle = \delta_{p,p'} \delta_{\sigma,\sigma'} N_{p\sigma} \langle P_{k-p-q,k,\sigma}(t_1) P_{k'-p',k'-q',\sigma'}^\dagger(t_2) \rangle = \delta_{\sigma,\sigma'} \delta_{k-p-q,k'-p'} \delta_{k,k'-q'} e^{-i\Delta\epsilon_{k,k-p-q}(t_1-t_2)} e^{-\gamma|t_1-t_2|}. \quad (13)$$

Working with this expression it is straightforward to get for $t' > t$ and steady state $t-t_0 \gg \gamma^{-1}, \gamma_s^{-1}$:

$$\langle S_z(q,t)S_z(q',t') \rangle^{(2)} = |d|^2 \delta_{q,-q'} \sum_{k,p} N_p \left[\frac{e^{-i(\Omega_{k,p,q} + \gamma)(t'-t)}}{\gamma_s^2 - (\gamma + i\Omega_{k,p,q})^2} - \frac{\gamma}{\gamma_s} \frac{e^{-\gamma_s(t'-t)}}{(\gamma_s - i\Omega_{k,p,q})^2 - \gamma^2} \right], \quad (14)$$

where $N_p = \sum_{\sigma} N_{p,\sigma}$ and $\Omega_{k,p,q} = \omega_p - \epsilon_k^c + \epsilon_{k-p-q}^v$. Note that since d is the interaction matrix element of the light-matter interaction, it scales like $V^{-1/2}$ because of the electric-field normalization. Therefore the correlations scale with the volume as V and are dimensionless (with our definition of S_z). It is instructive to study the structure of the expression inside the brackets in Eq. (14). First we note that it is stationary since the electronic correlations in Eq. (13) and the optical fields are also stationary. For a fixed k and p we expect naturally for the frequency $\Omega_{k,p,q}$ to appear with $t-t'$ (the first term). However, we also expect a buildup of a constant carrier density in the conduction band, which carries an inevitable static spin-density fluctuation $\langle S_z^2 \rangle$. Therefore we expect the spin fluctuation to have a dc Fourier component (the second term), balanced by the spin relaxation γ_s , and this indeed will

turn out to be the case when we evaluate the k summation (see below Sec. V).

IV. EFFECTS OF SQUEEZING

Let us now turn to the fourth order contribution to the correlations. Using expression (C1), we first average over the electronic $T=0$ ground state of the semiconductor (denoted by $\langle \cdot \rangle_{el}$). This averaging can be done on $S_z(q,t)^{(2)}$ independent of $S_z(q',t')^{(2)}$ since for $T=0$ there are no connected parts between the operators $n_{k,k'}^{c,v}$ in $\langle S_z(q,t)^{(2)} S_z(q',t')^{(2)} \rangle$. This gives, for steady state ($t-t_0 \gg \gamma^{-1}, \gamma_s^{-1}$):

$$\langle S_z(q,t) \rangle_{el} = |d|^2 \sum_{k,p} [\mathcal{S}_{k,p,\sigma=\uparrow} - \mathcal{S}_{k,p,\sigma=\downarrow}], \quad (15)$$

with

$$S_{k,p,\sigma} = \frac{e^{-i(\omega_p - \omega_{p-q})t} b_{p-q,\sigma}^* b_{p,\sigma}}{[\gamma_s - i(\omega_p - \omega_{p-q})][\gamma - i(\Delta\epsilon_{k-q,k-p} - \omega_{p-q})]} + \frac{e^{i(\omega_p - \omega_{p+q})t} b_{p,\sigma}^* b_{p+q,\sigma}}{[\gamma_s + i(\omega_p - \omega_{p+q})][\gamma + i(\Delta\epsilon_{k,k-p-q} - \omega_{p+q})]}, \quad (16)$$

where we introduce γ, γ_s already at the operator level as in the Heisenberg-Langevin approach, this being mathematically equivalent to adding them at the end as representing a single-particle level broadening and spin-wave frequency broadening. For the spin correlations we have the following dependence on fourth order correlations of the photon fields ($\langle \cdot \rangle$ denotes full averaging)

$$\begin{aligned} & \langle S_z(q,t) S_z(q',t') \rangle^{(4)} \\ &= |d|^4 \sum_{k,p} \sum_{k',p',\lambda} \left\{ \frac{e^{-i(\omega_p - \omega_{p-q})t} e^{-i(\omega_{p'} - \omega_{p'-q'})t'} [\langle b_{p-q,\lambda}^* b_{p,\lambda} b_{p'-q',\lambda}^* b_{p',\lambda} \rangle - \langle b_{p-q,\lambda}^* b_{p,\lambda} b_{p'-q',\lambda}^* b_{p',\lambda} \rangle]}{[\gamma_s - i(\omega_p - \omega_{p-q})][\gamma - i(\Delta\epsilon_{k-q,k-p} - \omega_{p-q})][\gamma_s - i(\omega_{p'} - \omega_{p'-q'})][\gamma - i(\Delta\epsilon_{k'-q',k'-p'} - \omega_{p'-q'})]} \right. \\ &+ \frac{e^{i(\omega_p - \omega_{p+q})t} e^{i(\omega_{p'} - \omega_{p'+q'})t'} [\langle b_{p,\lambda}^* b_{p+q,\lambda} b_{p',\lambda}^* b_{p'+q',\lambda} \rangle - \langle b_{p,\lambda}^* b_{p+q,\lambda} b_{p',\lambda}^* b_{p'+q',\lambda} \rangle]}{[\gamma_s + i(\omega_p - \omega_{p+q})][\gamma + i(\Delta\epsilon_{k,k-p-q} - \omega_{p+q})][\gamma_s + i(\omega_{p'} - \omega_{p'+q'})][\gamma + i(\Delta\epsilon_{k',k'-p'-q'} - \omega_{p'+q'})]} \\ &+ \frac{e^{-i(\omega_p - \omega_{p-q})t} e^{i(\omega_{p'} - \omega_{p'+q'})t'} [\langle b_{p-q,\lambda}^* b_{p,\lambda} b_{p',\lambda}^* b_{p'+q',\lambda} \rangle - \langle b_{p-q,\lambda}^* b_{p,\lambda} b_{p',\lambda}^* b_{p'+q',\lambda} \rangle]}{[\gamma_s - i(\omega_p - \omega_{p-q})][\gamma - i(\Delta\epsilon_{k-q,k-p} - \omega_{p-q})][\gamma_s + i(\omega_{p'} - \omega_{p'+q'})][\gamma + i(\Delta\epsilon_{k',k'-p'-q'} - \omega_{p'+q'})]} \\ &+ \left. \frac{e^{i(\omega_p - \omega_{p+q})t} e^{-i(\omega_{p'} - \omega_{p'-q'})t'} [\langle b_{p,\lambda}^* b_{p+q,\lambda} b_{p'-q',\lambda}^* b_{p',\lambda} \rangle - \langle b_{p,\lambda}^* b_{p+q,\lambda} b_{p'-q',\lambda}^* b_{p',\lambda} \rangle]}{[\gamma_s + i(\omega_p - \omega_{p+q})][\gamma + i(\Delta\epsilon_{k,k-p-q} - \omega_{p+q})][\gamma_s - i(\omega_{p'} - \omega_{p'-q'})][\gamma - i(\Delta\epsilon_{k'-q',k'-p'} - \omega_{p'-q'})]} \right\}, \quad (17) \end{aligned}$$

where each photon correlator can be factorized into products using intensity $\langle b_{q_1,\lambda_1}^* b_{q_1,\lambda_1} \rangle$ and squeezing $\langle b_{q_2,\lambda_2} b_{q_2,\lambda_2} \rangle$ since the field distribution is Gaussian for light which is down-converted using a $\chi^{(2)}$ nonlinearity.³⁹ As in the case of static spin correlations,²⁹ we assume a symmetric distribution of the correlations as functions of wave vectors, i.e., $q+q' = 2p_0$, which holds for nondegenerate (broad spectrum) down-converted light.⁴⁰ We can write for the correlations

$$\begin{aligned} \langle b_{p-q,\uparrow}^* b_{p,\uparrow} b_{p'+q',\uparrow}^* b_{p',\uparrow} \rangle &= N_{p-q} N_p \delta_{q,q'} \delta_{p,p'} \\ &+ M_{p-q}^{(1)*} M_p^{(1)} \delta_{q,q'} \delta_{p'+p-q,2p_0}, \\ \langle b_{p-q,\uparrow}^* b_{p,\uparrow} b_{p'+q',\uparrow}^* b_{p',\downarrow} \rangle &= N_{p-q} N_p \delta_{q,q'} \delta_{p-q,p'} \\ &+ M_{p-q}^{(1)*} M_p^{(1)} \delta_{q,q'} \delta_{p'+p,2p_0}, \\ \langle b_{p,\uparrow}^* b_{p+q,\uparrow} b_{p'+q',\uparrow}^* b_{p'-q',\uparrow} \rangle &= N_p N_{p+q} \delta_{q,q'} \delta_{p+q,p'} \\ &+ M_p^{(1)*} M_{p+q}^{(1)} \delta_{q,q'} \delta_{p'+p,2p_0}, \\ \langle b_{p,\uparrow}^* b_{p+q,\uparrow} b_{p'+q',\uparrow}^* b_{p',\downarrow} \rangle &= N_p N_{p+q} \delta_{q,q'} \delta_{p',p} \\ &+ M_p^{(1)*} M_{p+q}^{(1)} \delta_{q,q'} \delta_{p'+p+q,2p_0}, \end{aligned} \quad (18)$$

where p_0 is the central wave vector of the optical spectrum, and note we omitted the \pm index assuming $M_{q+}^{(\alpha)} = M_{q-}^{(\alpha)}$ for simplicity. In Fig. 2 we can see a diagrammatic representa-

tion of the microscopic processes that lead to the “ N ” and “ M ” terms in expression (18). These are essentially two particle-hole excitations which are correlated through the existence of photonic correlations. These photon correlations are spectral functions, which can be externally controlled at the source.^{26–28} Given these functions it is possible to characterize the fluctuations of the Stokes vector, whose average is assumed to be zero in this calculation (unpolarized).²⁹

We note that there is also a $\langle S_z(q,t) S_z(q',t') \rangle^{(3)}$ contribution which we discuss in detail in Appendix C. This contribution has a much smaller phase space compared to $\langle S_z(q,t) S_z(q',t') \rangle^{(2)}$ and therefore we neglect it. Also, we have shown³⁵ that a direct diagrammatic evaluation of the fourth order diagrams, Fig. 2, done in the Keldysh two-time formalism gives a result similar to expression (17), with the only difference being the replacement $\gamma_s \rightarrow \gamma$. The reason for

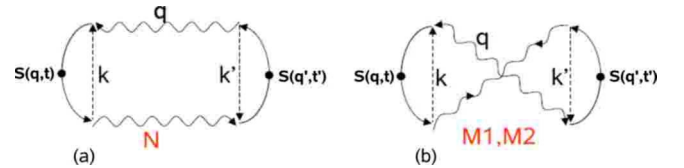


FIG. 2. (Color online) A diagrammatic representation of the resonant interaction processes in the normal and anomalous channels leading to spin correlations. (a) The normal channel is related to the autocorrelation function N of optical modes with themselves, whereas (b) the anomalous channel involves a correlation between different optical modes, either of same polarization (M_1) or of opposite polarization (M_2).

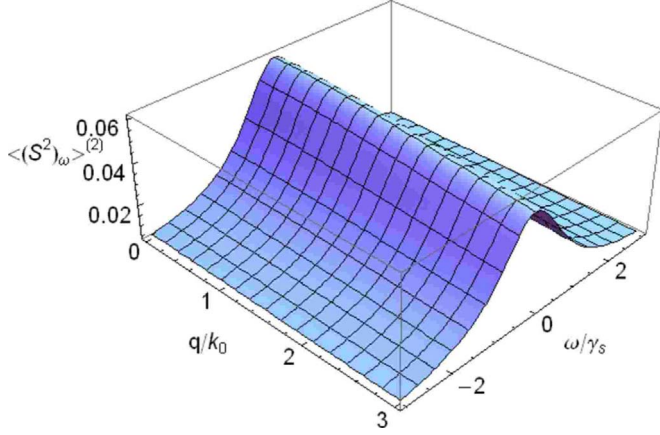


FIG. 3. (Color online) The spin-fluctuation spectrum (second-order contribution) plotted as a function of (q, ω) for excitations with a wave vector parallel to a unidirectional beam, scaled with the central electronic wave vector k_0 and the spin-relaxation rate γ_s , respectively.

this is that in the microscopic calculation we have not taken into account explicitly the spin-relaxation mechanisms, which is a topic for further research.

V. DISCUSSION AND RESULTS

We would like now to discuss and simplify the expressions for the spin-spin correlations obtained above. We begin from the second-order contribution (14) which is quite a complicated integral when the vector nature of k, p, q is taken into account. First note that for the problem we have in mind $p \ll k$, since typically $p \sim 1 \mu\text{m}^{-1}$ whereas $k_0 \sim 30 \mu\text{m}^{-1}$ for electrons with kinetic energy of 30 meV. In Fig. 3 results of numerical evaluation are shown for the time Fourier transform of Eq. (14), assuming a unidirectional beam $\vec{p} \parallel \hat{z}$ and spin-wave vectors in the direction of the beam $\vec{q} \parallel \vec{p}$. We see that the q dependence is very slow for $q \ll k_0$, and similarly it can be shown that the integrand dependence on p is negligible for $p \ll k_0$. Therefore a reasonable zeroth

approximation would be to neglect p and q altogether with respect to k . If we further restrict ourselves to small spin-wave momenta $q < B/c$ (B is the optical bandwidth in units of frequency), then $q \ll p$ and certainly this approximation is valid. We will also assume a constant electronic density of states in the energy range $\Omega = \pm \gamma$ where the integrand is appreciable and get a simple expression for the spin-spin correlations (*per unit volume*, and restoring missing \hbar 's)

$$\langle S_z(q, t) S_z(q', t') \rangle^{(2)} = \delta_{q, -q'} \frac{\pi |d|^2 \rho_{\text{el}} \sum_p N_p}{\hbar \gamma_s} e^{-\gamma_s |t-t'|}, \quad (19)$$

where $\rho_{\text{el}}(\epsilon_0) = \sqrt{2m^3 \epsilon / \hbar^3} |_{\epsilon = \epsilon_0}$ is the electron-hole density of states per unit volume in the bulk, taken at the central energy of excited electrons $\epsilon_0 = \hbar \omega_0 - E_g$, with m the reduced e - h effective mass and ϵ the excess kinetic energy of the e - h pair above the gap. Denoting $\langle (S_z^2)_{q, \omega} \rangle = F.T. [\langle S_z(q, t) S_z(q', t') \rangle]$ we obtain

$$\begin{aligned} \langle (S_z^2)_{q, \omega} \rangle^{(2)} &= \frac{2\pi |d|^2 \rho_{\text{el}} \sum_p N_p}{\hbar (\omega^2 + \gamma_s^2)} \\ &= \frac{2\pi |d|^2 \rho_{\text{el}} \int d\omega' \rho_{\text{opt}}(\omega') N(\omega')}{\hbar (\omega^2 + \gamma_s^2)}, \quad (20) \end{aligned}$$

where $N(\omega') \equiv N_q |_{\omega' = cq}$ is the photon distribution function in frequency space and $\rho_{\text{opt}}(\omega) = \omega^2 / \pi^2 c^3$ is the free space optical density of states per unit volume.⁴⁷ The contribution for long wavelengths is thus a Lorentzian background whose width is determined by the spin-relaxation time.

Turning now to the fourth order contribution, we observe that in certain physical situations it, Eq. (17), can be simplified considerably. This happens if the excess energy of the light above the electron-hole gap ($\hbar \omega_0 - E_g$) is large compared to the exciton binding energy. In this regime, using the correlations (18) and assuming that $k \gg p, q$, we get for the only important contribution the following approximate simple expression

$$\begin{aligned} \langle S_z(q, t) S_z(q', t') \rangle^{(4)} &= \delta_{q, -q'} \frac{2\pi^2 \rho_{\text{el}}^2 |d|^4 e^{-i\omega_q(t-t')}}{\hbar^2 (\omega_q^2 + \gamma_s^2)} \sum_p [N_{p-q, +} N_{p, +} + N_{p, +} N_{p+q, +} + N_{p-q, -} N_{p, -} + N_{p, -} N_{p+q, -} \\ &\quad + M_{p-q, +}^{(1)*} M_{p, +}^{(1)} + M_{p, +}^{(1)*} M_{p+q, +}^{(1)} \\ &\quad + M_{p-q, -}^{(1)*} M_{p, -}^{(1)} + M_{p, -}^{(1)*} M_{p+q, -}^{(1)} - M_{p-q, +}^{(2)*} M_{p, +}^{(2)} - M_{p-q, -}^{(2)*} M_{p, -}^{(2)} \\ &\quad - M_{p, +}^{(2)*} M_{p+q, +}^{(2)} - M_{p, -}^{(2)*} M_{p+q, -}^{(2)}], \quad (21) \end{aligned}$$

where ρ_{el} is the electron-hole joint density of states at $\Delta \epsilon_k = \hbar \omega_0$. Since the field is unpolarized $N_+ = N_-$, and assuming again for simplicity the symmetric case $M_{q+}^{(\alpha)} = M_{q-}^{(\alpha)} = M_q^{(\alpha)}$, to which we always refer from this point on. Therefore we can omit the polarization indices

$$\langle S_z(q, t) S_z(q', t') \rangle^{(4)} = \delta_{q, -q'} \frac{4\pi^2 \rho_{\text{el}}^2 |d|^4 e^{-i\omega_q(t-t')}}{\hbar^2 (\omega_q^2 + \gamma_s^2)} \sum_p [N_{p-q} N_p + N_p N_{p+q} + M_{p-q}^{(1)*} M_p^{(1)} + M_p^{(1)*} M_{p+q}^{(1)} - M_{p-q}^{(2)*} M_p^{(2)} - M_p^{(2)*} M_{p+q}^{(2)}]. \quad (22)$$

The restriction $q' = -q$ which reflects translational invariance leads to $\langle S_z(q, t) S_z(q', t') \rangle = \langle S_z(q, t') S_z(q', t) \rangle^*$. The right hand side of Eq. (22) indeed obeys this if the summation over p is real, which can be shown explicitly by using the symmetry property $M_p = M_{2p_0-p}$ of the squeezing correlation.

It is useful at this point to compare the decay and oscillatory time dependences of Eqs. (19) and (21), respectively. The second-order contribution (19) describes part of the spin fluctuations which is only due to the effect of the environment. The spin remains temporally correlated over a time scale τ_s that it takes for the relaxation processes to be effective. As can be seen from Fig. 4, the two orders of the interaction with the light are not sufficient to affect the dynamics of the correlation function $\langle S_z(q, t) S_z(q', t') \rangle$. The contribution of the fourth order processes is physically different. Here the spin-correlation dynamics reflects directly the correlations of the polarization of the light. This can be understood also diagrammatically from Fig. 2. In the fourth order contribution there is no electronic propagator temporally connecting $S_z(q, t)$ and $S_z(q', t')$ (see Fig. 2). Only the correlation functions of the light are connecting the two electronic diagrams, and the dynamics of the light is affecting the two-time dynamics. We see that the spin relaxation γ_s does affect the weight of each spectral component $S_z(q, t)$. Fast spectral components $\omega_q \gg \gamma_s$ are independent of γ_s and their weight is $\propto \omega_q^{-2}$, i.e., fast oscillations do not have enough time to be affected by the spin relaxation. In contrast, slow spectral components $\omega_q \ll \gamma_s$ are building up slow enough to

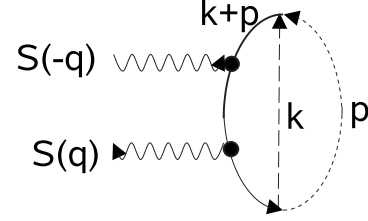


FIG. 4. Diagram of the second-order contribution to the spin-spin correlations. The electron in the valence band (dashed line) is excited into the conduction band (solid line) by the photon correlation (dotted line). The only possible dynamics between the times t, t' in this order of perturbation theory is due to the phenomenological spin relaxation (τ_s).

be balanced by γ_s and their weight is approximately $\propto \gamma_s^{-2}$. In particular, without the spin relaxation in the model we would have a divergence of the $q=0$ component, which corresponds to an unbalanced accumulation of spin-correlated charges in the conduction band due to the double absorption process. Note that there are also other diagrams of the fourth order where γ_s can affect the time dependence, however they have a much smaller contribution due to electronic phase-space considerations (see discussion in Appendix C and elsewhere²⁹).

The spectral function looks like

$$\langle (S_z^2)_{q, \omega} \rangle^{(4)} = \frac{4\pi^2 \rho_{\text{el}}^2 |d|^4 \delta(\omega - cq)}{\hbar^2 (\omega^2 + \gamma_s^2)} \sum_p [N_{p-q} N_p + N_p N_{p+q} + M_{p-q}^{(1)*} M_p^{(1)} + M_p^{(1)*} M_{p+q}^{(1)} - M_{p-q}^{(2)*} M_p^{(2)} - M_p^{(2)*} M_{p+q}^{(2)}]. \quad (23)$$

The δ function reflects the dispersion of the light. The p summation can be transformed to a frequency integral (so now the left hand side is *per unit volume*)

$$\langle (S_z^2)_{q, \omega} \rangle^{(4)} = \frac{4\pi^2 \rho_{\text{el}}^2 |d|^4 \delta(\omega - \omega_q)}{\hbar^2 (\omega^2 + \gamma_s^2)} \int d\omega' \rho_{\text{opt}}(\omega') [N(\omega' - \omega) N(\omega') + N(\omega') N(\omega' + \omega) + M^{(1)*}(\omega' - \omega) M^{(1)}(\omega') + M^{(1)*}(\omega') M^{(1)}(\omega' + \omega) - M^{(2)*}(\omega' - \omega) M^{(2)}(\omega') - M^{(2)*}(\omega') M^{(2)}(\omega' + \omega)]. \quad (24)$$

We see that as the phases of $M^{(1,2)}$ are modulated as a function of frequency, the spectrum of the driven spin fluctuations is also modulated. The spin structure factor in these situations is in fact *coherently controlled* using the phases of the ω -dependent squeezing. Combining the second (20) and fourth (24) order contributions, defining

$$C(\omega) = \int d\omega' [N(\omega' - \omega) N(\omega') + N(\omega') N(\omega' + \omega) + M^{(1)*}(\omega' - \omega) M^{(1)}(\omega') + M^{(1)*}(\omega') M^{(1)}(\omega' + \omega) - M^{(2)*}(\omega' - \omega) M^{(2)}(\omega') - M^{(2)*}(\omega') M^{(2)}(\omega' + \omega)] \quad (25)$$

and taking the optical density of states as approximately constant, given by $\rho_{\text{opt}} = \rho_{\text{opt}}(\omega_0)$ from the middle of the spectrum, we obtain

$$\langle (S_z^2)_{q, \omega} \rangle^{(2+4)} = \frac{2\pi |d|^2 \rho_{\text{el}} \rho_{\text{opt}}}{\hbar (\omega^2 + \gamma_s^2)} \left[\int d\omega' N(\omega') + 4\pi^2 \frac{\rho_{\text{el}} |d|^2}{\hbar} \delta(\omega - cq) C(\omega) \right]. \quad (26)$$

Note that this result is valid for long wavelengths ($q \ll B/c$), which is also the range where the spin structure factor is largest.

Let us now choose as direction of light propagation \hat{z} and integrate over q_z , setting $q_x = q_y = 0$. Since our approximations are only valid for small q , we will use a cutoff B/c on the q_z integration, which means that the result is the ω spectrum spatially averaged over the coherence length c/B in z direc-

tion and the x - y plane. Physically, this is motivated by having in mind a Faraday probe beam passing through the sample and measuring the spin fluctuations. For small frequencies $\omega \ll B$ this integration will smear out the δ -function singularity. We assume that the sample is much smaller compared to the coherence length of the light l_c , so the averaging is effectively done over the whole sample. This integration gives the following spin spectral density:

$$\langle (S_z^2)_{\omega} \rangle^{(2+4)} = \int_{-B/c}^{B/c} dq_z \langle (S_z^2)_{q,w} \rangle^{(2+4)}|_{q_x=q_y=0} = \frac{4\pi |d|^2 \rho_{\text{el}} \rho_{\text{opt}} B}{\hbar c (\omega^2 + \gamma_s^2)} \left[\int d\omega' N(\omega') + \frac{4\pi^2 \rho_{\text{el}} |d|^2}{\hbar B} C(\omega) \right]. \quad (27)$$

In order to estimate the strength of the coherent effects compared to the background of second-order fluctuations we need to calculate the ratio of the fourth order to the second order

$$\mathcal{R}(\omega) = \frac{4\pi^2 \rho_{\text{el}} d^2}{\hbar B} \frac{C(\omega)}{\int d\omega' N(\omega')}, \quad (28)$$

which at $\omega=0$ can be approximately estimated for strong classical squeezing (e.g., $M^{(1)}=N$) to give $16\pi^2 \rho_{\text{el}} d^2 \bar{N} / \hbar B$, where \bar{N} is the average photon occupation inside the optical bandwidth. Since this is a perturbative calculation, we expect the result, Eq. (27), to be valid as long as the second term is much smaller than the first one. This will generally put a restriction on the intensity, or photon occupation function $N(\omega)$, for a given optical bandwidth B . For an optical excitation which generates conduction-band electrons with kinetic energy of about 30 meV, and an optical energy of approximately 1.5 eV, we can estimate that for an optical bandwidth $\hbar B \approx 10$ meV the average photon occupation should be $10^{-4} - 10^{-5}$ for the second term to be 1/10 of the first one [$\mathcal{R}(\omega=0)=0.1$]. Usually if one uses correlated photons from down-converted light, the spectrum is very wide in the nondegenerate case, and therefore the average photon number per mode (\bar{N}) is very small. For example as discussed in Wang *et al.*⁴⁸ for type-I down-converted light, a counting rate of 10^4 photons/s was reported over a bandwidth of 10^{12} Hz, which corresponds to $\bar{N}=10^{-8}$. In more recent experiments, Bowen *et al.*,⁴⁹ Heersink *et al.*,²⁸ and Marquardt *et al.*⁵⁰ reported higher intensities for polarization-squeezed light.

It is interesting to note that according to the above estimates there appears to exist an experimentally accessible regime where the perturbative approximation breaks down ($N \geq \hbar B / 16\pi^2 \rho_{\text{el}} d^2$). In this regime higher orders in the light-matter interaction become important, leading to a strong nonlinear response to the driving field. It is an intriguing direction for future research, especially since strong correlated light sources are becoming increasingly available.^{27,28} An-

other interesting direction to explore is related to effects of nonclassical squeezed light $|M\rangle > N$ on the spectrum, which we showed to have a unique effect on the static spin correlations.²⁹

Although unexplored in this work, light in the quantum optical regime may have a unique effect on the temporal spin fluctuations. In another reference²⁹ we showed that the total (static) spin fluctuation can be reduced when the polarization fluctuations of the light are below the shot-noise limit. We believe that it is therefore reasonable to expect similar reduction to show up as negative values in the spectrally resolved contribution (24). A calculation of the quantum optical regime requires treating the light as a quantum field and taking into account its dynamics up to the fourth order.

VI. EXAMPLES OF PHASE MODULATIONS

The quantity $C(\omega)$ appearing in Eq. (27) is sensitive to the squeezing phases, i.e., the phases of $M(\omega)$ [cf. Eq. (25)]. In the general case $\arg[M(\omega)]$ can be expanded in a power series so initially it makes sense to focus on the linear and quadratic dependences. First let us rewrite the spectral spin density (27) as

$$\int_0^{B/c} dq_z \langle (S_z^2)_{q,w} \rangle^{(2+4)}|_{q_x=q_y=0} = \alpha \left(\tilde{\omega}^2 + \left(\frac{\gamma_s}{B} \right)^2 \right)^{-1} [1 + \eta \mathcal{C}(\tilde{\omega})], \quad (29)$$

where $\tilde{\omega} = \omega/B$, $\alpha = \frac{2\pi \rho_{\text{opt}} d^2 \rho_{\text{el}}}{cB} \int d\omega' N(\omega')$, and $\eta = 4\pi^2 \frac{\rho_{\text{el}}}{\rho_{\text{opt}}} \frac{\gamma_{\text{rec}}}{B} \bar{N}$, with the electron-hole recombination rate $\gamma_{\text{rec}} = \frac{2\pi}{3} \rho_{\text{opt}} d^2 / \hbar^2$, the average photon occupation \bar{N} , and $\mathcal{C}(\tilde{\omega}) = B^{-1} \bar{N}^{-2} C(B\tilde{\omega})$.

In the linear case we take $\theta(\omega - \omega_0) = T|\omega - \omega_0|$ with a phase modulation parameter $T = 2 \times 10^4$ s, small average photon number $\bar{N} = 10^{-4}$, optical bandwidth $\hbar B = 20$ meV, average electron kinetic energy $\epsilon_k = 30$ meV, and spin-relaxation time $\tau_s = 0.2$ ns. The photon correlation functions were taken to be Gaussian $N(\omega) = \exp\{-[(\omega - \omega_0)/B]^2\}$ and $M(\omega) = N(\omega) \exp[iT|\omega - \omega_0|]$. One possible experimental realization of a phase modulation of squeezed light is by using a pulse shaper.³⁴ With these parameters such a pulse shaper has

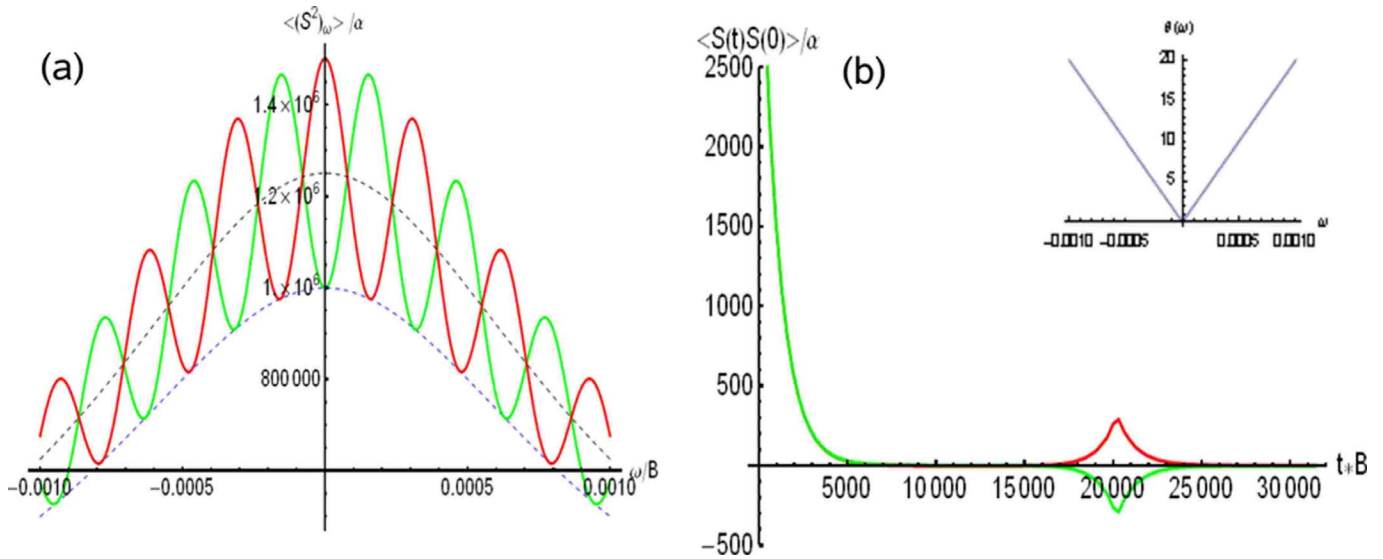


FIG. 5. (Color online) (a) ω dependence of the spin structure factor for a linear phase modulation of the squeezing phase of the field (insert: the dependence of the phase on the frequency). The figure depicts the second-order contribution (dashed blue) and phase-sensitive fourth order contributions for same-polarization (red) and opposite-polarization squeezing (green), respectively, compared to no squeezing (dashed black). (b) In the time domain we clearly see that the phase modulation parameter $T=20\,000$ s determines appearance time of the second correlation peak. The correlation between polarizations determine whether this peak will be positive (red) or negative (green).

to turn the squeezing phase a full cycle of 2π every 1.6 GHz. In Fig. 5(a) we draw the right hand side of Eq. (29) divided by α as a function of $\tilde{\omega}$. We see that the spectrum develops as additional oscillatory structure with a frequency T^{-1} , which depends on the type of squeezing (same or opposite polarization). The linear phase modulation in frequency space can be thought of as a time shift in the optical field in time domain [Fig. 5(b)]. This is somewhat similar to a retardation

of part of the random polarization signal, leading to the second rise of the spin correlation exactly after that time (T). This analogy is however not exact since the frequency shift is a symmetric function.¹¹

In the quadratic case (see Fig. 6), we take $\theta(\omega - \omega_0) = T(\omega - \omega_0)^2$ with a phase modulation parameter $T = 2 \times 10^4$ s², small average photon number $\bar{N} = 10^{-4}$, optical bandwidth $\hbar B = 20$ meV, and average electron kinetic en-

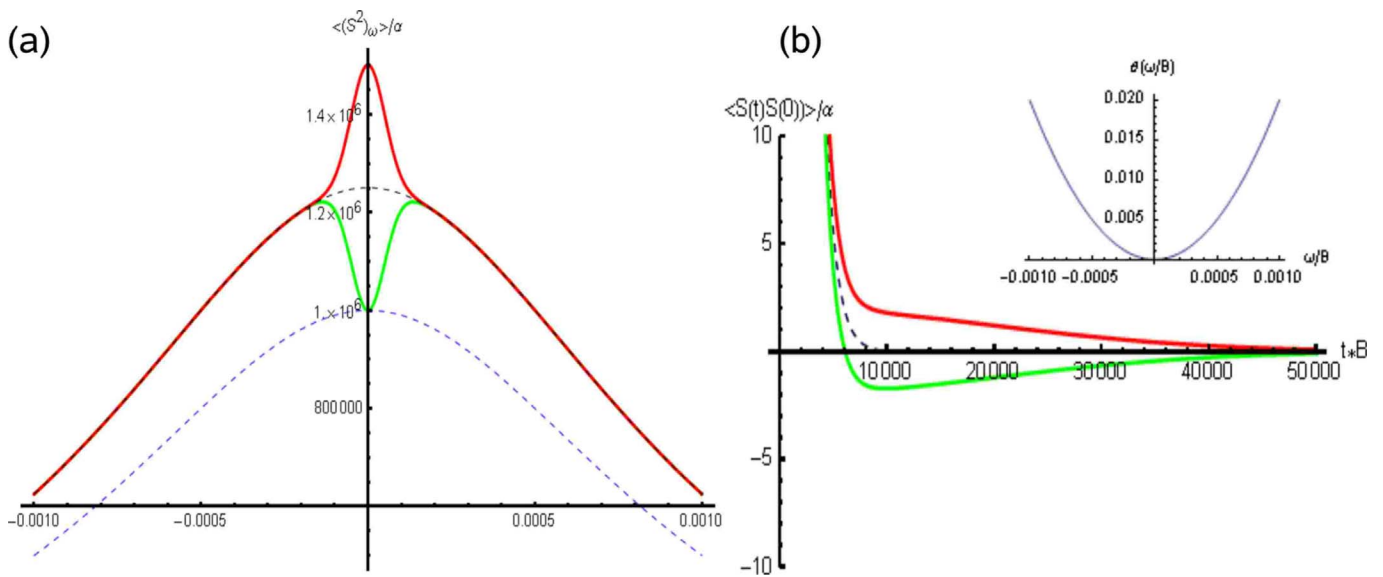


FIG. 6. (Color online) (a) ω dependence of the spin structure factor for a quadratic phase modulation of the squeezing phase of the field (insert: the dependence of the phase on the frequency). The figure depicts the second-order contribution (blue) and phase-sensitive fourth order contributions for same-polarization (red) and opposite-polarization squeezing, respectively, compared to no squeezing (green). (b) In time domain, in contrast to the linear case, the chirped phase modulation endows the spin correlation with a long tail of correlation determined by T rather than by τ_s and starting immediately after the time τ_s . Again the polarization correlation determines the sign of the long-range temporal correlation.

ergy $\epsilon_k=30$ meV. The photon correlation functions were taken to be again Gaussian with the squeezing $M(\omega)=N(\omega)\exp[iT(\omega-\omega_0)^2]$. In this case it is clear that the effect of “chirping” in ω space leads through the nonlinear formula Eq. (27) to an enhancement (reduction) of the correlations around the dc component of the spin correlations when the light is endowed with same (opposite) polarization correlations. In the time domain we see that there are two time scales, γ_s^{-1} and T , that govern the behavior of the spin correlations. For opposite-polarization correlations we observe a qualitative difference: on short-time scales $t < \gamma_s^{-1}$ the correlation is positive but for longer times $t > T$ it becomes negative. This change only happens in the opposite-polarization case when the squeezing phase is modulated on a scale much smaller than the spin-relaxation rate ($T^{-1} \ll \gamma_s$).

With the experimental parameters estimated above, $\eta \approx 0.1$ and $\alpha \approx 10^9$ s/m⁴. To estimate the strength of the total spin fluctuations we can look at the prefactor $\alpha/[\bar{\omega}^2 + (\gamma_s/B)^2]$ for $\bar{\omega}=0$ or at the integrated power, depending on what kind of experiment is performed. For the first case we get that the prefactor scales such as $\bar{N}B^2$, while $\eta \propto \bar{N}/B$, so the overall strength of the spin fluctuations can be increased while keeping η small. Since light with frequencies above the e - h gap is strongly absorbed with a typical attenuation coefficient of the order of 10^{-3} cm⁻¹ for GaAs, it is preferable to assume a thin slab, for example, with a thickness of $5 \mu\text{m}$ and a beam of light with an area of 1 mm^2 . For this geometry and the other parameters given above, the strength of the spin fluctuations around $\omega=0$ would be between 10 and 1000 in units of \hbar^2 , with the range reflecting an uncertainty of several parameters. This is the strength of the combined contribution of the second and fourth orders, so the signal (the fourth order) is estimated from η to be around 10%. Even though the total spin is very small for a macroscopic sample, we note that recently even a *single spin* has been measured with the Faraday rotation technique.⁵¹ In order to detect the features of the coherent modulation discussed here, it is necessary to sample the Faraday signal at least every B^{-1} seconds and also to detect slow changes of the order of T at the same time. In the amplitude domain, it is necessary to detect changes of the order of a few percent in the signal, as can be seen from Fig. 5.

VII. CONCLUSIONS

We calculated the steady-state spin fluctuations for a two-band semiconductor model driven by external light that con-

tains polarization correlations. We find for the spin structure factor a sensitivity to the phases of external optical correlations that can lead to time delays and oscillations. We believe that this application can be useful in optospintronics for additional control of spins beyond optical orientation. Additional interesting directions for research are the quantum optical regime, so far only explored for static spin correlations, and the regime of strong optical fields, which require a non-perturbative approach to the light-matter interaction.

APPENDIX A: OPTICAL TRANSITION MATRIX ELEMENTS

We summarize here for convenience the calculation of the dipole interaction matrix elements $d_{k,p}^s$ that appear in Eq. (2) for optical transitions in direct-gap semiconductors. In the spherical version of the Luttinger-Kohn model³⁶ for degenerate valence bands we can choose the angular-momentum quantization axis \hat{z} to be parallel to the optical beam \hat{n} . This model takes into account the spin-orbit interaction in the valence band, which leads to mixing of the *heavy-hole* (HH) and *light-hole* (LH) states ($j=3/2$). For the fourth order correlation processes which we consider, the strongest contributions from LH \rightarrow C transitions are still weaker by a factor of $1/3$. For this reason and for simplicity we consider only HH \rightarrow C in our model. Let us start with a given electronic wave vector k near the Γ point in the valence band. The wave function can be written as¹

$$\psi_{k,m}^v = e^{ikr} \sum_{\mu} \chi_{m\mu}(k) u_{\mu}^v, \quad (\text{A1})$$

where $\mu, m = \pm 3/2$ denote the HH eigenstates, $\chi_{m\mu}(k) = \mathcal{D}_{\mu,m}^{(3/2)}(\varphi, \theta, \phi)$ are rotation matrices⁵² (θ, φ are polar angles of \hat{k} , ϕ a global phase), and u_{μ}^v are angular-momentum eigenfunctions of J_z . The quantum number $m = \pm 3/2$ denotes the angular-momentum projection in the direction of \hat{k} and $\mu = \pm 3/2$ is the projection in the direction z . Using the usual notation for the wave functions of s and p orbitals, $u_{3/2}^v = -\frac{1}{\sqrt{2}}(X+iY)\uparrow$ and $u_{-3/2}^v = \frac{1}{\sqrt{2}}(X-iY)\downarrow$. For the conduction band $\psi_{k,s}^c = e^{ikr} u_s$, where $u_{1/2}^c = S\uparrow$ and $u_{-1/2}^c = S\downarrow$. The electronic parts of the dipole matrix elements are then given by

$$\langle \psi_{k,s}^c | \vec{D} | \psi_{k',m}^v \rangle = \delta_{k,k'} \sum_{\mu} \chi_{m\mu}(k) \langle u_s^c | \vec{D} | u_{\mu}^v \rangle, \quad (\text{A2})$$

where $\vec{D} = e\vec{r}$ is the dipole moment operator. Let us consider the matrix elements for the transition into the state $k, s = 1/2$:

$$\begin{aligned} \langle \psi_{k,1/2}^c | \vec{D} | \psi_{k',3/2}^v \rangle &= \delta_{k,k'} \left[\chi_{+3/2,+3/2}(k) \left\langle S\uparrow | \vec{D} | -\frac{1}{\sqrt{2}}(X+iY)\uparrow \right\rangle + \chi_{+3/2,-3/2}(k) \left\langle S\uparrow | \vec{D} | \frac{1}{\sqrt{2}}(X-iY)\downarrow \right\rangle \right] \\ &= \delta_{k,k'} \chi_{+3/2,+3/2}(k) \frac{-D}{\sqrt{2}} (\hat{x} + i\hat{y}), \end{aligned}$$

$$\begin{aligned} \langle \psi_{k,1/2}^c | \vec{D} | \psi_{k',-3/2}^v \rangle &= \delta_{k,k'} \left[\chi_{-3/2,+3/2}(k) \left\langle S \uparrow | \vec{D} | -\frac{1}{\sqrt{2}}(X+iY) \uparrow \right\rangle + \chi_{-3/2,-3/2}(k) \left\langle S \uparrow | \vec{D} | \frac{1}{\sqrt{2}}(X-iY) \downarrow \right\rangle \right] \\ &= \delta_{k,k'} \chi_{-3/2,+3/2}(k) \frac{-D}{\sqrt{2}} (\hat{x} + i\hat{y}), \end{aligned} \quad (\text{A3})$$

where we used the fact¹ that the only nonzero matrix elements are $\langle S | D_x | X \rangle = \langle S | D_y | Y \rangle = \langle S | D_z | Z \rangle = D$ due to the spatial symmetry of these integrals. We see that due to spin orthogonality only one of the orbital angular component $\mu = +3/2$ contributes to the transition into $+1/2$ state (a similar expression can be derived for $s = -1/2$). The full interaction matrix elements are given by $d_{k,k'-k}^{\lambda,s,m} = \mathcal{E}_{\omega_{k-k'}} \hat{\epsilon}_\lambda \cdot \langle \vec{D} \rangle_{k,k'}^{s,m}$, where $\lambda = \pm$ denotes the helicity and $\mathcal{E}_{\omega_{k-k'}} = \sqrt{\hbar \omega_{k-k'} / 2 \epsilon_0 V}$. We see that λ is determined by the conduction-band spin, i.e., $s \Leftrightarrow \lambda$, while for an arbitrary direction of \hat{k} both $m = \pm 3/2$ contribute to the transition. We can therefore omit the index λ and summation over it and continue with a simplified interaction Hamiltonian

$$\sum_{k,p} \sum_{s,m} d_{k,p}^{s,m} c_{k+p,s}^\dagger b_{p,s} v_{k,m} + \text{H.c.}, \quad (\text{A4})$$

where we now use s also for the helicity. By applying a transformation to the basis where angular-momentum projection is along \hat{n} with new operators $\tilde{v}_{k,\mu}$:

$$v_{k,m} = \chi_{m,+3/2}(k) \tilde{v}_{k,+3/2} + \chi_{m,-3/2}(k) \tilde{v}_{k,-3/2}, \quad (\text{A5})$$

the interaction appears as

$$\begin{aligned} &\sum_{k,p} \sum_{s,m} d_{k,p}^{s,m} c_{k+p,s}^\dagger b_{p,s} [\chi_{m,+3/2}(k) \tilde{v}_{k,+3/2} + \chi_{m,-3/2}(k) \tilde{v}_{k,-3/2}] + \text{H.c.} \\ &= \sum_{k,p} \sum_s c_{k+p,s}^\dagger b_{p,s} \{ [d_{k,p}^{s,+3/2} \chi_{+3/2,+3/2}(k) \tilde{v}_{k,+3/2} + d_{k,p}^{s,-3/2} \chi_{+3/2,-3/2}(k) \tilde{v}_{k,-3/2}] \\ &\quad + [d_{k,p}^{s,-3/2} \chi_{-3/2,+3/2}(k) \tilde{v}_{k,+3/2} + d_{k,p}^{s,+3/2} \chi_{-3/2,-3/2}(k) \tilde{v}_{k,-3/2}] \} + \text{H.c.} \\ &= \sum_{k,p} \sum_s c_{k+p,s}^\dagger b_{p,s} \{ [d_{k,p}^{s,+3/2} \chi_{+3/2,+3/2}(k) + d_{k,p}^{s,-3/2} \chi_{-3/2,+3/2}(k)] \tilde{v}_{k,+3/2} + [d_{k,p}^{s,+3/2} \chi_{+3/2,-3/2}(k) + d_{k,p}^{s,-3/2} \chi_{-3/2,-3/2}(k)] \tilde{v}_{k,-3/2} \} + \text{H.c.} \end{aligned} \quad (\text{A6})$$

The rotation matrix element can be written as⁵² $D_{m',m}^{(j)}(\varphi, \theta, \phi) = e^{im'\varphi} d_{m',m}^{(j)}(\theta) e^{im\phi}$ (here d is *not* the dipole matrix element). By using the symmetry relation $d_{m',m}^{(j)}(\theta) = (-1)^{m'-m} d_{-m',-m}^{(j)}(\theta)$ and setting $\phi = 0$, it is straightforward to show that the coefficient in front of $\tilde{v}_{k,+3/2}$ vanishes when $s = -1/2$ and the coefficient of $\tilde{v}_{k,-3/2}$ vanishes when $s = +1/2$. Therefore there is a one-to-one correspondence between the projection of angular momentum of the initial valence-band state μ and the projection of the spin of the final conduction-band state s . Renaming \tilde{v} as v , we can rewrite the interaction term as

$$\sum_{k,p} \sum_s c_{k+p,s}^\dagger b_{p,s} (-s) D \mathcal{E}_{\omega_p} [\chi_{s,s}^2(k) + \chi_{s,-s}^2(k)] v_{k,s} + \text{H.c.}, \quad (\text{A7})$$

where a common index $s = \pm 1 \Leftrightarrow \pm 1/2 \Leftrightarrow \pm 3/2$ is used for angular momentum, spin, and helicity. In cases where the angular dependence does not lead to other consequences other than some numerical prefactor (in cases where an isotropic approximation for all the other parts in the integrand is reasonable), we just omitted it and assume that D absorbs the prefactor.

APPENDIX B: PHENOMENOLOGICAL LEVEL WIDTHS

The fourth order contribution $\langle S(t)S(t') \rangle_{s,s}^{(4)}$ which we calculate here should match our previously calculated injection rate of static correlations²⁹ in the limit $t' \rightarrow t$. Indeed it can be shown that they give the same result if in the previous calculation a finite level width τ_s^{-1} is incorporated for the finite state propagator and the intermediate-state level width is taken to zero. To explain the source of this problem we need to compare the two approaches. Previously we calculated the static correlation ($t=t'$) directly using a second-order pertur-

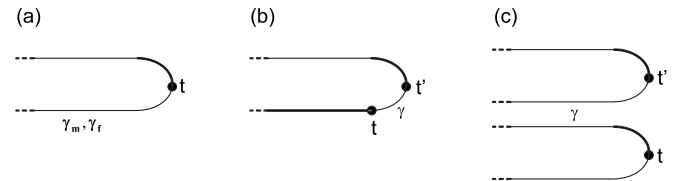


FIG. 7. A schematic for the time evolution for (a) direct evaluation of the static correlations via second-order perturbation theory. (b) The two-time spin-fluctuation function with a dissipative part shown in gray, and (c) the evaluation of the same correlation with equations of motion method.

bation expansion of the wave function. In interaction picture the static correlation can be written as [see Fig. 7(a)]

$$\langle\langle\psi_{\text{el}}|U^\dagger(t,-\infty)S(t)_0^2U(t,-\infty)|\psi_{\text{el}}\rangle\rangle_{\text{field}}, \quad (\text{B1})$$

while here we calculate the two-time correlation, which is defined as [see Fig. 7(b)]

$$\langle\langle\psi_{\text{el}}|U^\dagger(t,-\infty)S(t)_0U(t,t')S(t')_0U(t',-\infty)|\psi_{\text{el}}\rangle\rangle_{\text{field}}, \quad (\text{B2})$$

where $S(t)_0$ denote operators in the interaction picture. Since a direct evaluation of Eq. (B2) is very difficult for fourth order processes, we use the equations of motions for $S(t), S(t')$, which means actually that we split the intermediate time evolution into a product which gives [see Fig. 7(c)]

$$\langle\langle\psi_{\text{el}}|\tilde{U}^\dagger(t,-\infty)S(t)_0\tilde{U}(t,-\infty) \times \tilde{U}^\dagger(t',-\infty)S(t')_0\tilde{U}(t',-\infty)|\psi_{\text{el}}\rangle\rangle_{\text{field}}, \quad (\text{B3})$$

which is in principle equivalent to Eq. (B2) due to unitarity. The difficult issue is how to account also for the relaxation processes in such calculation in the simplest way while retaining consistency. In our calculations we added them explicitly as level broadenings (γ). However when the relaxation is added to the time evolution it is no longer possible to claim that unitarity applies

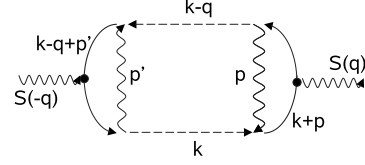


FIG. 8. Diagrammatic representation of the 1×3 excitation process with correlated photons. The diagram shows the emitted spin-wave (wiggly lines to the left and right of the diagram), pump light correlations (wiggly lines inside), and particle-hole excitations (solid dashed lines, respectively).

$$\tilde{U}(t,-\infty;\gamma)\tilde{U}^\dagger(t',-\infty;\gamma) \neq U(t,t';\gamma) \quad (\text{B4})$$

and therefore in principle different approaches can give different results. Specifically for the limit $t \rightarrow t'$ the evolution depicted in Fig. 7(a) might not give the same result as going along $(t \rightarrow -\infty \rightarrow t)$ in Fig. 7(c). In principle this shows that adding dissipations phenomenologically in higher order perturbation theory is very tricky, and it would be better to have a microscopic model instead. We have shown³⁵ another approach with Keldysh diagrammatic formalism that is promising to lead in the future to a derivation in which the dissipation processes will be derived from a microscopic Hamiltonian and will not suffer from such ambiguities.

APPENDIX C: CALCULATION OF THE $\langle S_z^{(1)}(q,t)S_z^{(3)}(q',t') \rangle$ CONTRIBUTION

We begin by writing explicitly the second contribution to the spin-wave operator

$$\begin{aligned} S_z(q,t)^{(2)} = & -|d|^2 e^{-\gamma t} \int_{t_0}^t dt_1 e^{-\gamma t_1} e^{\gamma t_1} \int_{t_0}^{t_1} dt_2 e^{\gamma t_2} \sum_{k,p,p'} \{ e^{i(\Delta\epsilon_{k-q,k-p-\omega_p})t_1} e^{-i(\Delta\epsilon_{k-q,k-p-\omega_{p'}})t_2} b_{p'\uparrow}^* [n_{k-q,k-p+p',\uparrow}^c(t_2)_0 - n_{k-q-p',k-p,\uparrow}^v(t_2)_0] b_{p\uparrow} \\ & + e^{-i(\Delta\epsilon_{k,k-p-q-\omega_p})t_1} e^{i(\Delta\epsilon_{k,k-p-q-\omega_{p'}})t_2} b_{p\uparrow}^* [n_{k-p-q+p',k,\uparrow}^c(t_2)_0 - n_{k-p-q,k-p',\uparrow}^v(t_2)_0] b_{p'\uparrow} - \\ & - e^{i(\Delta\epsilon_{k-q,k-p-\omega_p})t_1} e^{-i(\Delta\epsilon_{k-q,k-p-\omega_{p'}})t_2} b_{p'\downarrow}^* [n_{k-q,k-p+p',\downarrow}^c(t_2)_0 - n_{k-q-p',k-p,\downarrow}^v(t_2)_0] b_{p\downarrow} \\ & - e^{-i(\Delta\epsilon_{k,k-p-q-\omega_p})t_1} e^{i(\Delta\epsilon_{k,k-p-q-\omega_{p'}})t_2} b_{p\downarrow}^* [n_{k-p-q+p',k,\downarrow}^c(t_2)_0 - n_{k-p-q,k-p',\downarrow}^v(t_2)_0] b_{p'\downarrow} \}. \end{aligned} \quad (\text{C1})$$

In order to derive the third order contribution $S_z(q,t)^{(3)}$ it is necessary to formally solve the equations of motion for the various operators that appear in the second-order contribution (C1). We start by writing integral expressions for $n_{k,k'}^{c,v}$, again employing the basic equations of motion (7), and repeat the approximations of static spin waves by neglecting the free evolution of $n_{k,k'}$ terms

$$\begin{aligned} n_{k-q,k-p+p',\sigma}^c(t_2) = & e^{-\gamma t_2} \int_{t_0}^{t_2} dt_3 e^{\gamma t_3} i \sum_{p''} [-dP_{k-p+p'-p'',k-q,\sigma}^\dagger(t_3)_0 b_{p''\sigma}(t_3)_0 + d^* b_{p''\sigma}^*(t_3)_0 P_{k-q-p'',k-p+p',\sigma}(t_3)_0] \\ = & \sum_{p''} e^{-\gamma t_2} \int_{t_0}^{t_2} dt_3 e^{\gamma t_3} i [-e^{i(\Delta\epsilon_{k-q,k-p+p'-p''-\omega_{p''}})t_3} dP_{k-p+p'-p'',k-q,\sigma}^\dagger b_{p''\sigma} + e^{-i(\Delta\epsilon_{k-p+p',k-q-p''-\omega_{p''}})t_3} d^* b_{p''\sigma}^* P_{k-q-p'',k-p+p',\sigma}] \\ = & \sum_{p''} \left[\frac{-e^{i(\Delta\epsilon_{k-q,k-p+p'-p''-\omega_{p''}})t_2} dP_{k-p+p'-p'',k-q,\sigma}^\dagger b_{p''\sigma}}{\gamma + i(\Delta\epsilon_{k-q,k-p+p'-p''-\omega_{p''}})} + \frac{e^{-i(\Delta\epsilon_{k-p+p',k-q-p''-\omega_{p''}})t_2} d^* b_{p''\sigma}^* P_{k-q-p'',k-p+p',\sigma}}{\gamma - i(\Delta\epsilon_{k-p+p',k-q-p''-\omega_{p''}})} \right], \end{aligned} \quad (\text{C2})$$

$$\begin{aligned}
n_{k-p+p'-q,k,\sigma}^c(t_2) &= e^{-\gamma t_2} \int_{t_0}^{t_2} dt_3 e^{\gamma_3 t_3} \sum_{p''} [-dP_{k-p'',k-p+p'-q,\sigma}^\dagger(t_3)_0 b_{p''\sigma}(t_3)_0 + d^* b_{p''\sigma}^*(t_3)_0 P_{k-p+p'-p''-q,k,\sigma}(t_3)_0] \\
&= \sum_{p''} e^{-\gamma t_2} \int_{t_0}^{t_2} dt_3 e^{\gamma_3 t_3} i [-e^{i(\Delta\epsilon_{k-p+p'-q,k-p''-\omega_{p''}})t_3} dP_{k-p'',k-p+p'-q,\sigma}^\dagger b_{p''\sigma} + e^{-i(\Delta\epsilon_{k,k-p+p'-p''-q-\omega_{p''}})t_3} d^* b_{p''\sigma}^* P_{k-p+p'-p''-q,k,\sigma}] \\
&= \sum_{p''} \left[\frac{-e^{i(\Delta\epsilon_{k-p+p'-q,k-p''-\omega_{p''}})t_2} dP_{k-p'',k-p+p'-q,\sigma}^\dagger b_{p''\sigma}}{\gamma + i(\Delta\epsilon_{k-p+p'-q,k-p''-\omega_{p''}})} + \frac{e^{-i(\Delta\epsilon_{k,k-p+p'-p''-q-\omega_{p''}})t_2} d^* b_{p''\sigma}^* P_{k-p+p'-p''-q,k,\sigma}}{\gamma - i(\Delta\epsilon_{k,k-p+p'-p''-q-\omega_{p''}})} \right], \quad (C3)
\end{aligned}$$

$$\begin{aligned}
n_{k-p'-q,k-p,\sigma}^v(t_2) &= e^{-\gamma t_2} \int_{t_0}^{t_2} dt_3 e^{\gamma_3 t_3} \sum_{p''} [dP_{k-p',k-p+p'-q,p''\sigma}^\dagger(t_3)_0 b_{p''\sigma}(t_3)_0 - d^* b_{p''\sigma}^*(t_3)_0 P_{k-p'-q,k-p+p''\sigma}(t_3)_0] \\
&= \sum_{p''} e^{-\gamma t_2} \int_{t_0}^{t_2} dt_3 e^{\gamma_3 t_3} i [e^{i(\Delta\epsilon_{k-p'-q+p'',k-p-\omega_{p''}})t_3} dP_{k-p',k-p+p'-q,p''\sigma}^\dagger b_{p''\sigma} - e^{-i(\Delta\epsilon_{k-p+p'',k-p'-q-\omega_{p''}})t_3} d^* b_{p''\sigma}^* P_{k-p'-q,k-p+p''\sigma}] \\
&= \sum_{p''} \left[\frac{e^{i(\Delta\epsilon_{k-p'-q+p'',k-p-\omega_{p''}})t_2} dP_{k-p',k-p+p'-q,p''\sigma}^\dagger b_{p''\sigma}}{\gamma + i(\Delta\epsilon_{k-p'-q+p'',k-p-\omega_{p''}})} - \frac{e^{-i(\Delta\epsilon_{k-p+p'',k-p'-q-\omega_{p''}})t_2} d^* b_{p''\sigma}^* P_{k-p'-q,k-p+p''\sigma}}{\gamma - i(\Delta\epsilon_{k-p+p'',k-p'-q-\omega_{p''}})} \right], \quad (C4)
\end{aligned}$$

$$\begin{aligned}
n_{k-p-q,k-p',\sigma}^v(t_2) &= e^{-\gamma t_2} \int_{t_0}^{t_2} dt_3 e^{\gamma_3 t_3} \sum_{p''} [dP_{k-p',k-p+p''-q,\sigma}^\dagger(t_3)_0 b_{p''\sigma}(t_3)_0 - d^* b_{p''\sigma}^*(t_3)_0 P_{k-p-q,k-p'+p''\sigma}(t_3)_0] \\
&= \sum_{p''} e^{-\gamma t_2} \int_{t_0}^{t_2} dt_3 e^{\gamma_3 t_3} i [e^{i(\Delta\epsilon_{k-p+p''-q,k-p'-\omega_{p''}})t_3} dP_{k-p',k-p+p''-q,\sigma}^\dagger b_{p''\sigma} - e^{-i(\Delta\epsilon_{k-p'+p'',k-p-q-\omega_{p''}})t_3} d^* b_{p''\sigma}^* P_{k-p-q,k-p'+p''\sigma}] \\
&= \sum_{p''} \left[\frac{e^{i(\Delta\epsilon_{k-p+p''-q,k-p'-\omega_{p''}})t_2} dP_{k-p',k-p+p''-q,\sigma}^\dagger b_{p''\sigma}}{\gamma + i(\Delta\epsilon_{k-p+p''-q,k-p'-\omega_{p''}})} - \frac{e^{-i(\Delta\epsilon_{k-p'+p'',k-p-q-\omega_{p''}})t_2} d^* b_{p''\sigma}^* P_{k-p-q,k-p'+p''\sigma}}{\gamma - i(\Delta\epsilon_{k-p'+p'',k-p-q-\omega_{p''}})} \right]. \quad (C5)
\end{aligned}$$

When now substituting $n_{k,k}^{v,c}$ in the expression for $S_z^{(2)}(q,t)$, we only need the $\propto P^\dagger$ terms because in the correlation 1×3 only averages of the form $\langle PP^\dagger \rangle$ are contributing for zero temperature, leading to

$$\begin{aligned}
S_z(q,t)^{(3)} &= -|d|^2 e^{-\gamma t} \int_{t_0}^t dt_1 e^{-\gamma_1 t_1} e^{\gamma_2 t_1} \int_{t_0}^{t_1} dt_2 e^{\gamma_2 t_2} \sum_{k,p,p',p''} \left[e^{i(\Delta\epsilon_{k-p,k-q-\omega_p})t_1} e^{-i(\Delta\epsilon_{k-p,k-q-\omega_{p'}})t_2} b_{p'\uparrow}^* \right. \\
&\quad \times \left(\frac{-e^{i(\Delta\epsilon_{k-p+p',k-q-p''-\omega_{p''}})t_2} dP_{k-p+p'-p'',k-q,\uparrow}^\dagger b_{p''\uparrow}}{\gamma + i(\Delta\epsilon_{k-p+p',k-q-p''-\omega_{p''}})} - \frac{e^{i(\Delta\epsilon_{k-p+p'',k-p'-q-\omega_{p''}})t_2} dP_{k-p,k-p'-q+p''\uparrow}^\dagger b_{p''\uparrow}}{\gamma + i(\Delta\epsilon_{k-p+p'',k-p'-q-\omega_{p''}})} \right) b_{p\uparrow} \\
&\quad + e^{-i(\Delta\epsilon_{k-p-q,k-\omega_p})t_1} e^{i(\Delta\epsilon_{k-p-q,k-\omega_{p'}})t_2} b_{p\uparrow}^* \\
&\quad \times \left(\frac{-e^{i(\Delta\epsilon_{k-p+p'-q,k-p''-\omega_{p''}})t_2} dP_{k-p'',k-p+p'-q,\uparrow}^\dagger b_{p''\uparrow}}{\gamma + i(\Delta\epsilon_{k-p+p'-q,k-p''-\omega_{p''}})} - \frac{e^{i(\Delta\epsilon_{k-p+p''-q,k-p'-\omega_{p''}})t_2} dP_{k-p',k-p+p''-q,\uparrow}^\dagger b_{p''\uparrow}}{\gamma + i(\Delta\epsilon_{k-p+p''-q,k-p'-\omega_{p''}})} \right) b_{p'\uparrow} \\
&\quad - e^{i(\Delta\epsilon_{k-p,k-q-\omega_p})t_1} e^{-i(\Delta\epsilon_{k-p,k-q-\omega_{p'}})t_2} b_{p'\downarrow}^* \\
&\quad \times \left(\frac{-e^{i(\Delta\epsilon_{k-p+p',k-q-p''-\omega_{p''}})t_2} dP_{k-p+p'-p'',k-q,\downarrow}^\dagger b_{p''\downarrow}}{\gamma + i(\Delta\epsilon_{k-p+p',k-q-p''-\omega_{p''}})} - \frac{e^{i(\Delta\epsilon_{k-p+p'',k-p'-q-\omega_{p''}})t_2} dP_{k-p,k-p'-q+p''\downarrow}^\dagger b_{p''\downarrow}}{\gamma + i(\Delta\epsilon_{k-p+p'',k-p'-q-\omega_{p''}})} \right) b_{p\downarrow} \\
&\quad - e^{-i(\Delta\epsilon_{k-p-q,k-\omega_p})t_1} e^{i(\Delta\epsilon_{k-p-q,k-\omega_{p'}})t_2} b_{p\downarrow}^* \\
&\quad \times \left(\frac{-e^{i(\Delta\epsilon_{k-p+p'-q,k-p''-\omega_{p''}})t_2} dP_{k-p'',k-p+p'-q,\downarrow}^\dagger b_{p''\downarrow}}{\gamma + i(\Delta\epsilon_{k-p+p'-q,k-p''-\omega_{p''}})} - \frac{e^{i(\Delta\epsilon_{k-p+p''-q,k-p'-\omega_{p''}})t_2} dP_{k-p',k-p+p''-q,\downarrow}^\dagger b_{p''\downarrow}}{\gamma + i(\Delta\epsilon_{k-p+p''-q,k-p'-\omega_{p''}})} \right) b_{p'\downarrow} \left. \right]. \quad (C6)
\end{aligned}$$

The contraction $\langle S_z^{(1)}(\bar{q},t) S_z^{(3)}(q,t) \rangle$ leads to contractions such as

$$\langle b_{\bar{p}}^* b_{p'}^* b_{p''} b_p \rangle \langle P_{\bar{k}-\bar{p}-\bar{q}, \bar{k}}^- P_{k-p+p'-p'', k-q}^+ \rangle, \quad (C7)$$

which result in the constraint $\bar{k}=k-q$, which restrict the phase space of k, \bar{k} summation considerably compared to the 2×2 contribution. Diagrammatically, these fourth order contributions can be described as one fermion loop diagram (see Fig. 8).

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