Manipulation of collective spin correlations in semiconductors with polarization squeezed vacuum

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We calculate the transfer rate of correlations from polarization entangled photons to the collective spin of a many-electron state in a two-band system. It is shown that when a semiconductor absorbs pairs of photons from a two-mode squeezed vacuum, certain fourth order electron-photon processes correlate the spins of the excited electron pairs of different quasi-momenta. Different distributions of the quantum Stokes vector of the light lead to either enhancement or reduction of the collective spin correlations, depending on the symmetry of the distribution. We find that as the squeezing of the light becomes non-classical, the spin correlations exhibit a crossover from being positive with $N \sim N^2$ ($N$ is average photon number) scaling, to being negative with $\sim N$ scaling, even when $N$ is not small. Negative spin correlations mean a preponderance of spin singlets in the optically generated state. We discuss the possibility to measure the collective spin correlations in a combined measurement of the Faraday rotation fluctuation spectrum and excitation density in a steady-state configuration.

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Optical excitation of a semiconductor with circularly polarized light generates an average collective spin polarization in the conduction band. This allows to investigate spin relaxation mechanisms in semiconductors using techniques such as time-resolved Faraday rotation, time-resolved photoluminescence, and femtosecond pulses. It is also possible to monitor the position of the electron spins, their phase and amplitude as well as coherently control them. Recently the fluctuations of electronic collective spin of a bulk GaAs sample were measured. Theoretically, spin states are interesting because of their relation to squeezing and to entanglement. It has been suggested that collective atomic spins can store entanglement, optically transferred from correlated photons to the atomic cloud.

In this work we study how the spin correlations in a semiconductor system can be optically generated in a controlled way. We consider a pump beam of correlated photons which is absorbed in a semiconductor, exciting a density of electron-hole pairs. Absorbed photons are found to either enhance or reduce the spin-spin correlations depending on the correlations of optical modes with different wavelengths and either the same or different polarizations. In particular we find that it is necessary to use squeezed light in its non classical regime in order to excite net anti-correlated spins. This is due to a competition between two fourth-order processes inducing positive and negative correlations, the latter becoming dominant only for non-classical light. We note that net spin anticorrelations mean a preponderance of singlet spin components.

Polarization properties of photons are described by the quantum Stokes parameters which in the circular polarization basis ($\epsilon_{\pm}$) are written as, Ref. 22,

$$\hat{p}_{\pm} = (\epsilon_+ \cdot \mathbf{E}^\dagger - \epsilon_- \cdot \mathbf{E}^\dagger) \sigma_i \left( \begin{array}{c} \epsilon_+ \cdot \mathbf{E} \\ \epsilon_- \cdot \mathbf{E} \end{array} \right) = \sum_{q,q'} \hat{b}_{q}^\dagger \sigma_i \hat{b}_{q'}$$

where $\mathbf{E}$ is the electric field $\mathbf{E} = \sum_q \hat{\epsilon}_q b_{q\lambda}$ at position $r = 0$, $\hat{b}_q^\dagger = (b_{q,+}^\dagger, b_{q,-}^\dagger)$ and $\sigma_i = 0, 1, 2$ denote the unit and Pauli spin matrices. The operators $b_{q\lambda}$ are the photon annihilation operators with wave number $q$ and polarization $\lambda$. We consider a collinear pump beam with a range of frequencies $\omega_0 \pm B/2$ above the electron-hole gap. We assume that the average Stokes parameters in a given bandwidth $B$ are $\langle \hat{p}_0 \rangle = \pi c N \delta_{\lambda,\lambda'} \delta_{qq'}$, $\langle \hat{p}_{\pm} \rangle = \langle \hat{p}_z \rangle = 0$, where $N_{q\lambda}$ is the average photon occupation per mode and $L$ is the quantization length. This describes an unpolarized light with the Stokes vector.

The fluctuations are described by the covariance matrix $p_{ij} = 1/2(\hat{p}_i \hat{p}_j + \hat{p}_j \hat{p}_i)$, which for a Gaussian type field depends on the normal $\langle b_{q\lambda}^\dagger b_{q'\lambda'} \rangle$ as well as anomalous $\langle b_{q\lambda} b_{q'\lambda'} \rangle$ correlations, the latter constituting the main characteristics of squeezed vacuum. Beams with such properties are generated using the parametric down-conversion. In addition to normal correlations $\langle b_{q\lambda}^\dagger b_{q'\lambda'} \rangle = N_{q\lambda} \delta_{\lambda,\lambda'} \delta_{qq'}$ they possess two generic anomalous correlations: same polarization squeezing $\langle b_{q\lambda}^\dagger b_{q'\lambda'} \rangle = M_{q\lambda}^{(1)} \delta_{\lambda,\lambda'} \delta_{qq'}$, $\omega_{q,q'}$, $2\omega_0$, and opposite polarization squeezing $\langle b_{q\lambda}^\dagger b_{q'\lambda'}^\dagger \rangle = M_{q\lambda}^{(2)} \delta_{\lambda,\lambda'} \delta_{qq'}$, $\omega_{q,q'}$, $2\omega_0$, where $M_{q\lambda}^{(1,2)}$ are complex functions. Since for the squeezed vacuum $\langle \hat{p}_0 \hat{p}_{1,2,3} \rangle = 0$, the fluctuations of $\hat{p}_{1,2,3}$ can be described separately from the variance of $\hat{p}_0$.

It is instructive to draw the covariance ellipsoids for the tensor $p_{ij}$. Such ellipsoids are shown in Fig. 11 for the two cases of the same-polarization and opposite-polarization
squeezing based on averaged occupation and squeezing functions \( \hat{N}_k = N \) and \( M_k^{(1,2)} = M_k^{(1,2)} \). On the axes are plotted \( p_i \), possible values of the averages \( \langle \hat{p}_i \rangle \). The variance \( p_{33} \) given by \( 2[N(N + 1) + |M^{(1)}|^2 \pm |M^{(2)}|^2] \) for \( |M_k^{(1,2)}| = |M_k^{(1,2)}| \) indicating that correlations of the type \( M^{(1)} \) (\( M^{(2)} \)) enhance (reduce) the variance \( p_{33} \), a fact which is important for spin-spin correlations.

The free part of the Hamiltonian of the semiconductor is modelled as a two-band system

\[
H_0 = \sum_{q, \lambda} \omega_q b_{q\lambda}^\dagger b_{q\lambda} + \sum_{k, \sigma} \varepsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k, \sigma} \varepsilon_{k\sigma} v_{k\sigma}^\dagger v_{k\sigma}. \tag{2}
\]

The operators \( c_{k\sigma} \) and \( v_{k\sigma} \) denote annihilation operators of the free electrons in the conduction and valence bands, with quasi-momentum \( k \) and spin \( \sigma \). The interaction Hamiltonian of the electrons and the photons in the dipole approximation is given by

\[
V = \sum_{\sigma, \sigma', \lambda} \sum_{p, q} A_{pq}^{\sigma\sigma', \lambda \lambda'} \lambda \lambda' (p_{\sigma}^+ p_{\sigma'}^+ v_{\sigma'} v_{\sigma'} + h.c.)
\]

where \( A_{pq}^{\sigma\sigma', \lambda \lambda'} \) are the interaction matrix elements for dipole transitions from heavy-hole band to conduction band in GaAs near the \( \Gamma \) point.\[1] \[\[\]

**FIG. 1:** Two generic fluctuation patterns of the Stokes vector \( p_1, p_2, p_3 \). In the circular basis (\( \pm \)), they are described by correlation ellipsoids for (a) opposite-polarization squeezing (for average parameters \( N = 1, M_1^{(1)} = 0, M_2^{(1,2)} = 1.314 \)) and (b) same-polarization squeezing (\( N = 1, M_1^{(1)} = 1.314, M_2^{(1)} = 0 \)). The value of \( M \) was chosen to be slightly below the maximal squeezing \( \sqrt{N(N+1)} \).

In the present work we will not consider the electron-electron and electron-phonon interactions. The quantum optical effects on the rate of generation which we will describe in the context of the simple two band model should qualitatively hold also in the presence of the interactions. We will make more detailed comments supporting this assumption later on.

For the optical beam described above we wish to calculate the rate at which it is generating collective spin correlations in a semiconductor. The average total spin \( \langle S \rangle \) of the photo-excited conduction electrons is zero since the squeezed vacuum radiation is unpolarized. Consider the spin-noise two time average

\[
\langle S(t) \cdot S(t') \rangle = \sum_{k, k'} \langle S_k(t) \cdot S_{k'}(t') \rangle \tag{3}
\]

where \( S_k = \sum_{\sigma} c_{k\sigma}^\dagger \sigma_{k\sigma}^\dagger c_{k\sigma} \). It describes the fluctuations of the total spin \( S \) and consists of two parts - the sum of fluctuations of individual spins \( \sum_k \langle S_k(t) \cdot S_k(t') \rangle \) and the collective pairwise correlations, i.e. terms with \( k \neq k' \). Their variance is given by \( \langle S^2 \rangle - \sum_k \langle S_k^2 \rangle \).

To lowest order in the dipole interaction the contribution to the collective correlations comes from the optically induced transition of two electrons into states \( k, \sigma \) and \( k', \sigma' \). Physically, we expect that the two excited spins will be positively (negatively) correlated if the two absorbed photons are correlated and have the same (opposite) polarizations. Indeed the main result, Eq. \( S \) below, of the calculation reflects the competition between the strengths of the correlations existing in the photon field: an auto-correlation within each mode, and a cross-correlation between different modes due to the squeezing.

The second order contribution for the total spin fluctuations is given by

\[
\langle S^2 \rangle^{(2)} = \sum_{\{1,4\}} A_{p_1, q_1}^{\sigma_1 \sigma_1', \lambda_1 \lambda_1'} A_{p_2, q_2}^{\sigma_2 \sigma_2', \lambda_2 \lambda_2'} C_{p_1, q_1}^{\sigma_1 \sigma_1', \lambda_1 \lambda_1'} C_{p_2, q_2}^{\sigma_2 \sigma_2', \lambda_2 \lambda_2'} \times \langle b_{q_1}^{\sigma_1', \lambda_1} b_{q_2}^{\sigma_2', \lambda_2} b_{q_3} \rangle_{\text{rad}} \langle 1, 2 | S^2 | 3, 4 \rangle_{\text{eq}} \tag{4}
\]

where \( \langle b_{q_1}^{\sigma_1', \lambda_1} b_{q_2}^{\sigma_2', \lambda_2} b_{q_3} \rangle_{\text{rad}} \) is a property of the external field, and we define

\[
C_{p_1, q_1}^{\sigma_1 \sigma_1', \lambda_1 \lambda_1'} = \frac{\varepsilon^{2t} \varepsilon^{\lambda_1 \lambda_1'} \varepsilon^{\lambda_2 \lambda_2'} - \varepsilon^{\lambda_2 \lambda_2'} \varepsilon^{\lambda_1 \lambda_1'}}{\varepsilon^{\lambda_1 \lambda_1'} \varepsilon^{\lambda_2 \lambda_2'} - \varepsilon^{\lambda_2 \lambda_2'} \varepsilon^{\lambda_1 \lambda_1'}} \times \left( \frac{1}{\varepsilon^{\lambda_1 \lambda_1'}} - \varepsilon^{\lambda_2 \lambda_2'} \frac{1}{\varepsilon^{\lambda_1 \lambda_1'}} \right) \tag{5}
\]

which is the second order amplitude, where \( \varepsilon^{\sigma \sigma'} \) is the adiabatic switching on factor, and \( \varepsilon^\lambda \) is the lifetime of the conduction electron state. \( \varepsilon_{1}, \varepsilon_{2} \) is the fermionic average

\[
\langle 1, 2 | S^2 | 3, 4 \rangle_{\text{eq}} \tag{6}
\]

A Wick decomposition of expression (4) contains contractions which contribute to the independent fluctuations \( \sum_k \langle S_k^2 \rangle \) as well as contractions which contribute to the collective spin-spin correlations \( \langle S_k \cdot S_{k'} \rangle \) with \( k \neq k' \). The latter can be further divided into two processes, Fig. 2., in which (1) a singlet \( (k, k') \) in the valence
band is broken into two different momenta in the conduction band \((k + q, k + q')\) and (ii) two electrons with different momenta \((k_1, k_2)\) are excited into the conduction band with momenta \((k_1 + q, k_2 + q')\). Process (i) has considerably smaller rate with respect to (ii) because most of the phase space of final states cannot be reached with the typically small photon momentum. The ratio can be approximately estimated to be \((\frac{B}{q})^2\) where \(B\) is the optical bandwidth, and \(k\) is the typical electron wave number. Therefore in the following we neglect the contribution of process (i).

For the generation rate of correlations due to process (ii), we use the corresponding contractions in (11), differentiating with respect to time and taking the limit \(\eta \to 0\). This results in an energy conservation constraint for the entire process of exciting two electron-hole pairs. This process again has two parts (Fig. 2b): one coming from normal contractions \(\langle bb\rangle\) and another from anomalous contractions \(\langle bb'\rangle\) due to squeezing. For normal contractions any two spin components \(S_k, S_{k'}\) become correlated due to the absorption of two photons from the same mode \(q\), obeying the energy conservation \(\omega_q = \frac{1}{2}(\Delta \epsilon_k + \Delta \epsilon_{k'})\). In contrast, for anomalous contractions only spin components which have symmetric energies \(\Delta \epsilon_k + \Delta \epsilon_{k'} = 2\omega_0\) become correlated. These are drawn out of a continuum of such pairs obeying \(\omega_q + \omega_{q'} = 2\omega_0\). Therefore the two processes are distributed very differently in phase space, although they have the same total phase space. These processes give the largest contribution to the generation rate of correlations \((S^2) - \sum_k (S_k^2)\), and in the limit of \(q \ll k\) are given by (per unit volume)

\[
C_s = C_{s0} \sum_q \int d\Delta \epsilon_k \rho(\Delta \epsilon_k) \rho(2\omega_0 - \Delta \epsilon_k) \times \frac{\gamma^2}{(\Delta \epsilon_k - \omega_q)^2 + (\frac{\gamma}{2})^2} \left( N_q^2 + |M_q^{(1)}|^2 - |M_q^{(2)}|^2 \right)^2
\]

where \(C_{s0} = \frac{32\pi^3 |d|^4}{3\hbar}\) with \(d\) the dipole matrix element, and we assume \(|M_q^{(1,2)}| = |M_q^{(1,2)}|\) i.e. that the \((\pm)\) squeezing correlations differ only by phase. The numerical factors in \(C_{s0}\) is due to angular integrals and a symmetry factor of the contraction. We see from expression (7) that away from the edges of the bandwidth the correlation per unit bandwidth is simply proportional to \(N^2 + |M^{(1)}|^2 - |M^{(2)}|^2\) (assuming \(N_q \approx N\) and \(M_q^{(1,2)} \approx M^{(1,2)}\) across most of the bandwidth). Since the electronic density of state in the excitation bandwidth can be taken as constant, integrating (7) with respect to the electronic energy \(\Delta \epsilon_k\) gives

\[
C_s = C_{s0} \rho(\omega_0)^2 \frac{4 \pi}{\gamma} \sum_q \left( N_q^2 + |M_q^{(1)}|^2 - |M_q^{(2)}|^2 \right)^2.
\]

This result shows that increased (decreased) fluctuations of the Stokes parameter \(p_3\) cf. Fig 1 lead to increased (decreased) spin correlations. Positive spin correlations induced by absorbing photons from the same mode are enhanced by absorbing squeezed photons with the same polarization modes and reduced by the squeezing of the opposite polarizations, cf. Fig. 3. The total spin correlations can become negative when the squeezing is non-classical, i.e. \(M^{(2)} > N\). The maximal negative spin correlations will be reached for a pure squeezed state with \(M^{(2)} = \sqrt{N(N+1)}\), Ref. 27, in which case \(C_s \sim -N\). Remarkably this is a rare case in which a quantum optical effect is not confined to small photon occupations \(N \ll 1\).
It is instructive to consider the equal spin correlation curves, Fig. 3 which indicate that the geometry of negative correlation curves is separated from the other cases: the hyperbolic curves are confined to either the classical side ($C_\alpha > 0$) or the quantum side ($C_\alpha < 0$) of the diagram, with the $C_\alpha = 0$ being a separatrix between the two regimes. The possibility to completely eliminate the inevitable spin correlations induced by unsqueezed light (the first term in Eq. (8)) may be useful for observing other sources of spin correlations such as contributions from nuclear spins.

It is useful to define a reduced density matrix $\rho_{\alpha\beta}^{(k,k')}$ for a pair of spins $S_k, S_k'$, where $\alpha, \beta$ run over the singlet $\langle |0,0\rangle$ and three triplet basis states $\langle |1,0\rangle, |1,\pm 1\rangle$. In this basis it can be easily shown that for negative spin correlations $C_\alpha < 0$ the diagonal elements $\rho_{\alpha\alpha}^{(k,k')}$ obey

$$\sum_{k,k'} \rho_{0,0}^{(k,k')} > \frac{1}{3} \sum_{k,k'} [\rho_{1,-1}^{(k,k')} + \rho_{1,0}^{(k,k')} + \rho_{1,1}^{(k,k')}]$$

which means that for the electronic state generated by non-classical light, there is a preponderance of the singlet component in the pairwise spin correlations.

An enhancement or reduction of spin-spin correlations should be measurable from the difference of $\langle S^2 \rangle$ with squeezed and unsqueezed light, as can be seen from Eq. (3). It is also in principle possible to observe the spin correlations by measuring $\langle S^2 \rangle$ and $\sum_k \langle S_k^2 \rangle$. The total spin fluctuations $\langle S^2 \rangle$ can be estimated from the variance of the magnetic moment of the sample for example in a Faraday rotation setup similar to the one used to measure thermal spin fluctuations $\mathbb{R}$. For the diagonal part of the fluctuations we can use the identity

$$\sum_k \langle S_k^2 \rangle = 3 \sum_k \langle n_k^2 \rangle - 6 \sum_k \langle n_k^2 n_k \rangle$$

where the average $\langle n_k^2 n_k \rangle$ can be well approximated by $\langle n_k^2 \rangle \langle n_k \rangle$ since the correlated part of second order processes creating singlets at the same $k$ is very small compared to first order contribution. Therefore the knowledge of $\langle n_k \rangle$, e.g. from measurement of the excitation density can yield the information necessary for the estimation of $\sum_k \langle S_k^2 \rangle$.

Electron spins are decorrelated by random spin flip processes in semiconductors. It should be advantageous to use samples with long spin lifetime, such as in n-type bulk GaAs. Spin flip times of the photo-excited holes are much faster compared to the electrons, and therefore their contribution to the collective spin correlations should be small.

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