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# Observation of spin beats at the Rabi frequency in microcavities

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Rabi oscillations are observed in the optically induced time-resolved Faraday rotation from a strong-coupling microcavity. They reveal the dynamics of the excitonic part of the polariton state, including the spin related information. The oscillations of the same period are observed at both circular and linear pumping, suggesting the splitting of the ground polariton state into a linearly polarized doublet.

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Any two-level quantum system can show quantum beats if a linear combination of its eigenstates is coherently excited. Rabi oscillations in microcavities[1] represent quantum beats in a mixed exciton-photon system. In the strong coupling regime,[2] the exciton-polariton eigenstates are split by an energy dependent on the exciton oscillator strength and detuning between exciton and cavity modes.[3] At zero detuning, this splitting is referred to as the vacuum field Rabi splitting. When excited by a short laser pulse, the polariton system is initially in a purely photonic state $|C\rangle$, so that both the upper polariton branch $|U\rangle=\frac{1}{\sqrt{2}}(|X\rangle+|C\rangle)$ and lower polariton branch $|L\rangle=\frac{1}{\sqrt{2}}(|X\rangle-|C\rangle)$ are coherently excited, $|X\rangle$ being the pure exciton state. As time goes by, the system evolves according to $\frac{1}{\sqrt{2}}(|U\rangle e^{i\omega_U t}-|L\rangle e^{i\omega_L t})$, so that at $t=\frac{\pi}{\omega_U-\omega_L}$ it passes to the purely excitonic state. Afterwards it becomes photonic again etc., with $\omega_L$, $\omega_U$ being frequencies of lower and upper polariton modes. The Rabi oscillations persist until the phase correlation between upper and lower polariton branches is lost. In other words, their decay time is governed by the coherence lifetime in the system, so that a high quality factor is prerequisite to observe Rabi oscillations. The oscillations can be detected most easily by measurement of the photonic component of the cavity polariton state by the time-resolved reflection.[1,4,5] The dynamics of the excitonic component of the polariton state can be measured experimentally by the Kerr rotation technique.[6] Detection of the Rabi oscillations by this technique is a nontrivial task, as the decoherence of the ensemble of excitons created in the system is quite fast: excitons efficiently scatter with acoustic phonons and with each other, which reduces the coherence.[7]

Here we report an experimental observation of the oscillations of the excitonic component of the polariton doublet in a microcavity. We were able to reveal the dynamics of the excitonic part of the polariton state using the time-resolved Faraday rotation technique. This is a two beams technique where two optical pulses, separated by a time delay $\Delta t$, are sent to the sample. The first pulse (pump) creates a population of polarized excitons. The second pulse (probe) is linearly polarized. After transmission, its polarization plane is rotated by an angle proportional to the total exciton spin projection onto the light propagation direction. In the absence of an external magnetic field, this projection usually decays exponentially because of spin depolarization and radiative recombination of excitons. We shall see now that in microcavities instead of the monotonous decay, the Faraday rotation angle shows extremely pronounced oscillations, which are caused by the beats between excitonic and photonic components of the polariton state. It turns out that by use of Rabi oscillations at zero magnetic field the magnetization in a semiconductor can be efficiently controlled on a picosecond scale! The effect we observe has a different nature from recently reported Rabi oscillations in a two-level system based on coupled electron spins in double quantum dots[8] which do not involve light-matter coupling.

We study a high quality λ GaAs microcavity containing a single InGaAs quantum well (QW) and surrounded by two $AlAs/Al_{0.1}Ga_{0.9}As$ Bragg mirrors with 28 (22) pairs on the substrate (air) side. The sample was wedge shaped, so that the detuning $\delta$ between photon and exciton modes can be controlled by the displacement of the 100 $\mu$m diameter laser spot on the surface of the sample (8.4 meV/mm). An acromatic lens was used to focus pump and probe beams on the surface, in order to avoid pulse front distortions. In the experiments below we choose $\delta\sim 0$, which yields the splitting between the polariton modes equal to a 3.3 meV Rabi splitting. The corresponding transmission spectrum is shown in Fig. 1, inset. Both pump and probe pulses are incident at small oblique angles opposite to each other, about +2 (−2) degrees with respect to the surface normal. The pump/probe intensity ratio is equal to 15, the pump beam intensity is set to 3.5 mW. The probe pulse is linearly polarized along the $x$ axis (an in-plane axis). Its rotation after transmission through the sample as a function of the pump-probe delay is obtained

 

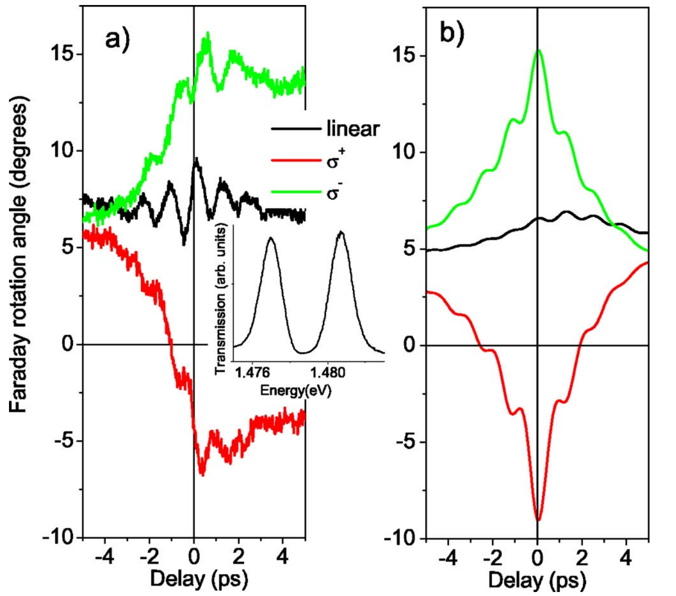


FIG. 1. (Color online) Faraday rotation of the probe pulse measured (a) and calculated (b) at the energy of low polariton state as a function of the pump-probe delay for $\sigma^+$, $\sigma^-$, and linearly ($X$) polarized pump pulse. The probe pulse is $X$ polarized. The inset shows the transmission spectrum at zero detuning.

for each pump polarization from the set of the three independent measurements. Namely, a linear polarizer is placed after the sample, and the intensity of the transmitted light for the three different orientations of the polarizer is measured using a monochromator at the energy of the low polariton branch $E_L = 1.4774$ eV. To get rid of the polarization effects on the monochromator diffraction efficiency, a 5 m multimode optical fibre is used to depolarize the light prior to detection. Note also that both linear and pump induced rotation of the probe polarization are detected in this configuration.

Figure 1(a) shows the time-resolved Faraday rotation measured for different polarizations of the pump pulse: $\sigma^+$ and $\sigma^-$ circular polarizations and $X$-linear polarization. All the spectra show pronounced oscillations with the period of about 1.25 ps, which corresponds to the energy splitting of about 3.3 meV, i.e. the value of Rabi splitting in our system. Without doubt, the observed oscillations result from the Rabi oscillations. This confirms that our microcavity is indeed in the strong coupling regime at the pumping intensity we use. We can also note that the Faraday effect in microcavities is *about 1000 times stronger* than in conventional quantum well structures due to accumulation of the polarization rotation angle during multiple round trips of light inside the cavity. The dramatic amplification of Faraday and Kerr effects in microcavities has been predicted in Ref. 9.

The other important feature of the Faraday rotation induced by the microcavity is the setup of the pump-induced rotation at negative pump-probe delays much longer than the duration of the pulse (150 fs). This is characteristic for microcavities, where the polaritons excited by the probe pulse live for some time in the cavity before escape, giving contribution to the Faraday rotation signal. If during their lifetime the circularly polarized polaritons are created by the pump pulse, the Faraday rotation can be induced.

Surprisingly, the Faraday rotation is nonzero at long negative time delays between pump and probe. This signal does not depend on the pump polarization and intensity and persists on the level of 6°–7° at very long positive and negative delays, i.e. in the absence of the pump effect. A strong Faraday-rotation signal can also be seen at linear pumping. The oscillations at linear pumping have the same period as the beats at circular pumping, but slightly different phase. The curves in Figure 1 showing the rotation angles measured for the two different circular polarizations of the pump are only slightly asymmetrical with respect to the value of signal in the absence of the pump. We show here that these effects are caused by the small splitting of the ground polariton state into a linearly polarized doublet.

The dynamics of polarized polaritons created by pump or probe pulses can be described using the generalized Liouville equations for the polariton density matrices at the pump and probe states $\rho^{(0)}$ and $\rho^{(1)}$, respectively:

$$i\hbar\frac{\partial\rho^{(0)}}{\partial t} = [H_0,\rho^{(0)}] - L\rho^{(0)} + P_{pump}(t), \tag{1}$$

$$i\hbar\frac{\partial\rho^{(1)}}{\partial t} = [H_0 + H_{ex}(\rho^{(0)}),\rho^{(1)}] - L\rho^{(1)} + P_{probe}(t), \tag{2}$$

where

$$H_0 = \begin{bmatrix} E_x & 0 & \frac{1}{2}V_R & 0 \\ 0 & E_x & 0 & \frac{1}{2}V_R \\ \frac{1}{2}V_R & 0 & E_{ph} & \Omega \\ 0 & \frac{1}{2}V_R & \Omega^* & E_{ph} \end{bmatrix} \tag{3}$$

is the linear Hamiltonian of the QW microcavity written in the basis of the two circularly polarized exciton and two circularly polarized photon states, $E_X$ is the heavy hole exciton energy, $E_{ph}$ is the photon energy, and $V_R$ is the Rabi splitting. $|\Omega|$ is the effective splitting of the ground state of the lower exciton-polariton branch.The lowest energy polariton eigenstate is assumed to be polarized along $x'$ axis inclined by an angle $\varphi$ with respect to the $x$ axis, so that $\Omega = |\Omega| e^{i\varphi}$ [see Fig. 2(d)]. The linear modes splitting is commonly observed in II-VI semiconductor based microcavities[10] and recently has also been observed for a similar microcavity sample.[11] Our preliminary experiments on this sample suggest $\Omega = 0.2$ meV. Possible origins of this splitting have been analyzed in Ref. 10 with the conclusion that most probably it is due to the optical birefringence in the Bragg mirrors. Note also that the splitting resulting from long-range exchange interaction of polaritons localized by structural disorder potential was calculated in Ref. 12 and is too small to account for the observed polarization rotation. The nonlinear part of the Hamiltonian $H_{ex}(\rho)$ in Eq. (2) describes the blue shift of the exciton energies due to the exciton-exciton interactions:

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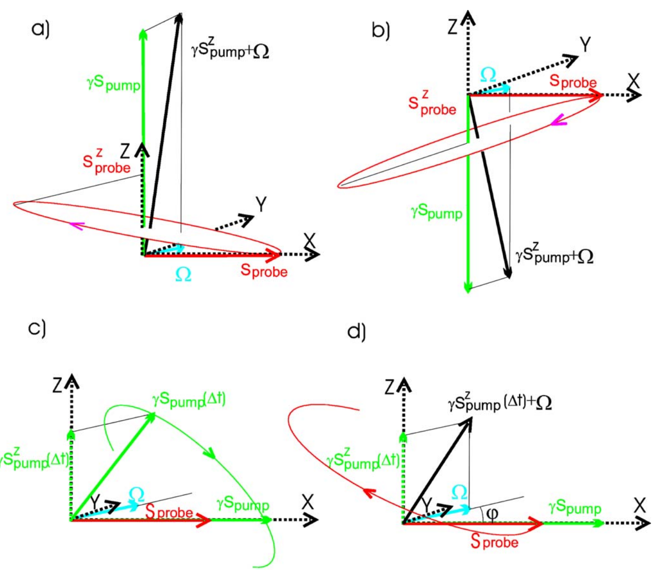


FIG. 2. (Color online) Pseudospin dynamics schemes for $\sigma^+$ (a), $\sigma^-$ (b), and linearly (c), (d) polarized pump pulse. The probe pseudospin is rotated around the vector sum of the in-plane field $\mathbf{\Omega}$ and the effective field proportional to the $Z$ component of the pump pseudospin. The pump pseudospin also precesses about the field $\mathbf{\Omega}$ [shown only for linear polarization in (c)], and gains the $z$ component which influences the probe pseudospin rotation (d)].

$$H_{ex}(\rho) = \begin{bmatrix} \gamma\rho_{11} & 0 & 0 & 0 \\ 0 & \gamma\rho_{22} & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}. \quad (4)$$

Here, $\gamma$ can be estimated as $\gamma \approx \frac{6E_B a_B^2}{S} \approx 5\times 10^{-8}$ meV, where $E_B$ is an exciton binding energy, $a_B$ is a two-dimensional (2D) exciton Bohr radius, $S$ is the laser spot area.[13,14] Another source of nonlinearity in the system could be the dependence of the coupling constant $V_R$ on the exciton concentration. Such a dependence comes from the exciton oscillator strength reduction due to the phase space filling. It was shown theoretically that this nonlinearity may give the contribution of the same order of magnitude as the exciton-exciton interaction.[13,14] Therefore, we have checked numerically introducing the corresponding terms in the Hamiltonian (4), that the phase space filling nolinearity results in the oscillation in Kerr rotation with much lower contrast than that induced by the exciton blue shift. In addition, from Ref. 15 follows that the phase space filling effects should play a minor role with respect to the Coulomb interaction effects in our experimental conditions. Thus these terms are neglected in the calculations below.

The operators $\hat{L}$ and $\hat{P}_{pump,probe}$ account for the polariton relaxation and generation, respectively. The dissipation of the elements of the density matrix due to the radiative decay and decoherence processes is described by Lindblad terms:

$$\hat{L}\rho = i\hbar \begin{bmatrix} \rho_{11}/\tau_{ex} & \rho_{12}/\tau_{ex} & \rho_{13}/\tau & \rho_{14}/\tau \\ \rho_{21}/\tau_{ex} & \rho_{22}/\tau_{ex} & \rho_{23}/\tau & \rho_{24}/\tau \\ \rho_{31}/\tau & \rho_{32}/\tau & \rho_{33}/\tau_{ph} & \rho_{34}/\tau_{ph} \\ \rho_{41}/\tau & \rho_{42}/\tau & \rho_{43}/\tau_{ph} & \rho_{44}/\tau_{ph} \end{bmatrix}, \quad (5)$$

where $\tau_{ex}$ and $\tau_{ph}$ are exciton and photon lifetimes, respectively, and $\frac{1}{\tau}=\frac{1}{\tau_{ex}}+\frac{1}{\tau_{ph}}$. Here we assume that the loss of coherence is due to the finite polariton lifetime only. The excitation of polaritons in the system is described by terms:

$$P_{pump}(t) = i\hbar \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & f_{pump}(t) & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}, \quad (6)$$

for the circularly polarized pump,

$$P_{probe}(t) = i\hbar \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & f_{probe}(t)/2 & f_{probe}(t)/2 \\ 0 & 0 & f_{probe}(t)/2 & f_{probe}(t)/2 \end{bmatrix} \quad (7)$$

for the linear polarized probe, where $f_{pump}(t) = N_{pump}\delta_p \exp(-\delta_p^2 t^2)$, $f_{probe}(t) = N_{probe}\delta_p \exp[-\delta_p^2(t-\Delta t)^2]$, $N_{pump(probe)}$ is proportional to the intensity of the pump (probe) pulse, $\Delta t$ is the time delay between the two pulses, $1/\delta_p$ is the pulse duration. If the pump is linearly polarized, $P_{pump}(t)$ is given by Eq. (7) where $f_{probe}$ should be substituted by $f_{pump}$.

In Eqs. (1) and (2), we take into account the nonlinear effect of the intense pump pulse on the weak probe pulse and neglect for simplicity all other nonlinearities (self-effects of the pump- and probe-probe effect on the pump state).

The angle of Faraday rotation is dependent on the ratio of the in-plane components of the pseudospin[16] of the photonic part of the cavity modes:

$$\varphi(\Delta t) = \frac{1}{2}\arctan\frac{S_y(\Delta t)}{S_x(\Delta t)}, \quad (8)$$

where

$$S_x(\Delta t) = \mathrm{Re}\left[\int_{-\infty}^{+\infty} \rho^{(1)}{}_{34}(t)dt\right],$$

$$S_y(\Delta t) = \mathrm{Im}\left[\int_{-\infty}^{+\infty} \rho^{(1)}{}_{34}(t)dt\right]. \quad (9)$$

Equations (8) and (9) describe the spectrally integrated optical response of the system. If the signal is detected only at the energy of one of the polariton branches, it is convenient to rewrite the density matrix of the system in the basis of spin-up and spin-down polarized upper and lower polariton states. If the signal is detected at the low-polariton branch energy, the matrix element of interest writes

$$\rho'_{34} = \beta_+\beta_-\rho_{12} + \alpha_+\alpha_-\rho_{34} - \alpha_-\beta_+\rho_{14} - \alpha_+\beta_-\rho_{32}. \quad (10)$$

The coefficients $\alpha_+$, $\alpha_-$, $\beta_+$, $\beta_-$ are elements of the normalized eigenvectors of the $2\times 2$ Hamiltonian matrices:

$$\begin{bmatrix} E_X^{\pm} & V_R \\ V_R & E_{ph} \end{bmatrix}. \tag{11}$$

In the case of nonzero detuning they are given by

$$\alpha_{\pm} = \frac{V_R}{\sqrt{2V_R^2 + \frac{1}{2}(E_{ph}-E_X^{\pm})^2 + (E_{ph}-E_X^{\pm})\sqrt{\left(\frac{E_{ph}-E_X^{\pm}}{2}\right)^2 + V_R^2}}}, \tag{12}$$

$$\beta_{\pm} = \left(\frac{E_{ph}-E_X^{\pm}}{2} + \sqrt{\left(\frac{E_{ph}-E_X^{\pm}}{2}\right)^2 + V_R^2}\right)\frac{\alpha_{\pm}}{V_R}. \tag{13}$$

The time dependence of the Faraday rotation signal for the low polariton branch is always given by Eqs. (8) and (9), where one has to substitute $\rho_{34}$ by $\rho'_{34}$.

In the numerical calculation we have used the following set of parameters: $E_X=E_{ph}=1.4774$ eV, $V_R=3.3$ meV, $N_{pump}=15N_{probe}=10^9$, $\gamma=5.5\times10^{-8}$ meV, $\tau_{ex}=100$ ps, $\tau_{ph}=2$ ps, $|\mathbf{\Omega}|=0.15$ meV, $\varphi=-18°$. Note, that here only $\Omega$ and $\tau_{ph}$ are fitting parameters, since all the others are known from cw experiments and $\tau_{ex}$ has very little effect on the signal decay time. Figure 1(b) shows the calculated time-resolved Faraday rotation for three experimental configurations. One can see a good agreement between theory and experiment. However, one can see that oscillations in the case of the linearly polarized excitation are more pronounced in the experiment than in the simulations, and the phase shift of all the curves with respect to zero pump-probe delay could not be described by the model. Additionally, under circularly polarized excitation, a slowly decaying Faraday rotation at positive times is observed. It is probably due to spin polarized exciton population created by the pump pulse and localized in the QW, which is not accounted for by the model. Nevertheless, the most important features, namely, Rabi oscillations, asymmetry between the spectra taken at $\sigma^+$ and $\sigma^-$ pump polarizations, oscillations at linear pumping, and their phase shift from the oscillations at circular pumping, the rotation of probe pulse in the absence of the pump are reproduced by the model. Qualitatively, the complex polarization dynamics of exciton polaritons in our experiment can be understood in terms of the probe pseudospin precession about the effective field created by the pump pulse along the $z$ axis and the in-plane effective field $\mathbf{\Omega}$ (see Fig. 2). The first kind of precession is a manifestation of the optically induced Faraday rotation. If only this effect would be present, the spectra at $\sigma^+$ and $\sigma^-$ pump would be symmetric and no probe polarization rotation at linear pump would be observed. Due to the in-plane field $\mathbf{\Omega}$, both pump and probe pseudospins experience an additional rotation with a period much longer than the Rabi oscillation period. This field breaks the symmetry of $\sigma^+$ and $\sigma^-$ pump experiments. In the case of linear pump, it leads to the buildup of the $z$ component of the pump pseudospin, which causes the in-plane rotation of the probe pseudospin. In the absence of the pump, the effective in-plane field induces the linear birefringence, which is responsible for the experimentally observed 6°–7° rotation signal at very long positive and negative delays between pump and probe pulses.

In conclusion, the spin beats due to Rabi oscillations have been observed in microcavities. The spin is transferred from excitons to photons and vice versa with the frequency given by the Rabi splitting of exciton-polariton modes. Our experiment has demonstrated the possibility of the optical control of the magnetization in a semiconductor on a time scale shorter than the exciton radiative lifetime.

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[1] T. B. Norris, J. K. Rhee, C. Y. Sung, Y. Arakawa, M. Nishioka, and C. Weisbuch, Phys. Rev. B **50**, 14663 (1994).

[2] R. Houdre, C. Weisbuch, R. P. Stanley, U. Oesterle, P. Pellandini, and M. Ilegems, Phys. Rev. Lett. **73**, 2043 (1994).

[3] V. Savona, L. C. Andreani, P. Schwendimann, and A. Quattropani, Solid State Commun. **93**, 733 (1995).

[4] J. D. Berger, O. Lyngnes, H. M. Gibbs, G. Khitrova, T. R. Nelson, E. K. Lindmark, A. V. Kavokin, M. A. Kaliteevski, and V. V. Zapasskii, Phys. Rev. B **54**, 1975 (1996).

[5] S. Jiang, S. Machida, Y. Takiguchi, Y. Yamamoto, and H. Cao, Appl. Phys. Lett. **73**, 3031 (1998).

[6] A. Brunetti, M. Vladimirova, D. Scalbert, R. André, D. Solnyshkov, G. Malpuech, I. A. Shelykh, and A. V. Kavokin, Phys. Rev. B **73**, 205337 (2006).

[7] G. Malpuech, Y. G. Rubo, F. P. Laussy, P. Bigenwald, and A. V. Kavokin, Semicond. Sci. Technol. **18**, S395 (2003).

[8] J. R. Petta, A. C. Johnson, J. M. Taylor, E. A. Laird, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Science **309**, 2180 (2005).

[9] A. V. Kavokin, M. R. Vladimirova, M. A. Kaliteevski, O. Lyngnes, J. D. Berger, H. M. Gibbs, and G. Khitrova, Phys. Rev. B **56**, 1087 (1997).

[10] J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. M. J. Keeling, F. M. Marchetti, M. H. Szymanska, R. André, J. L. Staehli, V. Savona, P. B. Littlewood, B. Deveaud, and Le Si Dang, Nature (London) **443**, 409 (2006); Ł. Klopotowski, M. D. Martin, A. Amo, L. Viña, I. A. Shelykh, M. M. Glazov, G. Malpuech, A. V. Kavokin, and R. André, Solid State Commun. **139**, 511 (2006).

[11] D. N. Krizhanovskii, D. Sanvitto, I. A. Shelykh, M. M. Glazov, G. Malpuech, D. D. Solnyshkov, A. Kavokin, S. Ceccarelli, M. S. Skolnick, and J. S. Roberts, Phys. Rev. B **73**, 073303 (2006).

[12] V. Savona, C. Piermarocchi, A. Quatropani, P. Schwendimann, and F. Tassone, Phase Transitions **68**, 169 (1999); V. Savona and F. Fassone, Solid State Commun. **95**, 673 (1995).

[13] F. Tassone and Y. Yamamoto, Phys. Rev. B **59**, 10830 (1999).

[14] C. Ciuti, P. Schwendimann, and A. Quatropani, Semicond. Sci. Technol. **18**, S279 (2003).

[15] A. Huynh, J. Tignon, Ph. Roussignol, C. Delalande, R. André, R. Romestain, and D. Le Si Dang, Phys. Rev. B **66**, 113301 (2002).

[16] Pseudospin is a quantum analog of the Stokes vector. Its $Z$ component describes the circular polarization degree, the $X$ and $Y$ components describe the linear polarization degree and orientation. More details are in Ref. 11.