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Polarized light bursts from kicked quantum rings

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Nonequilibrium quantum rings emit circular-polarized subterahertz radiation with a polarization degree controllable on nano- to picosecond time scales. This we conclude using a theory developed here for the time-dependent detection of the circular polarization of polychromatic radiations, valid for time scales comparable to the reciprocal of characteristic emission frequencies. The theory is applied to driven quantum rings whereby the influence of radiative and nonradiative processes on the properties of the emitted light is incorporated.

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Kekulé's work on the benzene molecule [1] initiated a continuing fascination with quantum rings. The Aharonov-Bohm effect, persistent charge, and spin currents are just but a few examples of the intriguing phenomena naturally emerging in these structures [2]. Much effort is currently devoted to light-driven molecular and nano- and mesoscopic rings [3–10], a development fueled by the spectacular advance in the controlled fabrication of these materials [11–16] paralleled by equally impressive progress in laser technology. Light irradiation may trigger nonequilibrium charge polarization and ring currents with associated ultrafast electric and magnetic pulse emission [3,5–10,17], which can be harnessed for applications, e.g., as a polarized light-pulse source emitting in a wide range of frequencies. In view of these prospects a thorough understanding of the time structure, polarization, and decay channels of the emitted light is mandatory. The aim of the current work is to show that polarization properties of the emitted radiation, especially the degree of the circular polarization, can be controlled on ultrafast time scales. For that, we developed a theory for the ultrafast, time-dependent detection of light polarization properties.

The radiation of electrons moving on a circular orbit was considered as early as 1907 by Schott in an attempt to explain discrete atomic spectra using classical electrodynamics [18]. His results, although not resolving the original problem, were reproduced and became important much later [19,20] in the context of synchrotron radiation; the polarization characteristics of the angular-resolved spectrum were analyzed [21]. Here we generalize these results to a time-dependent case using the theory of a time-dependent spectrum [22,23] and apply them to the emission from driven quantum rings that can serve as miniature light sources with a polarization switchable within picoseconds. Such a possibility to shape the polarization on ultrafast time scales, which are comparable to the reciprocal of characteristic emission frequencies, opens new perspectives for applications with respect to existing emitters of (linear or circular) polarized light based on quantum nanostructures [24]. The geometry of a quantum ring allows us to achieve both almost purely circular and linear polarized emission. In general, the time dependence of Stokes parameters can be shaped by appropriate driving fields.

For a driven quantum ring (with radius r_0) we inspect the emitted far-field radiation at a point $\mathbf{R}$, $R \gg r_0$. The far-field intensity per solid angle $\Omega=(\theta,\phi)$ follows from $\frac{dI}{d\Omega}=\frac{\sqrt{\kappa}}{4\pi c^3}|\langle\hat{\boldsymbol{\mathcal{E}}}(\mathbf{R},t)\rangle|^2$ (we use Gauss units), where κ is the medium dielectric constant, c is the light speed, $\hat{\boldsymbol{\mathcal{E}}}$ is the electric field operator, and $\langle\cdots\rangle$ denotes the expectation value. Extending the theory of a time-dependent spectrum [22,23] to the polarized case we find the detected time t and frequency ω dependence of the intensity $I(\omega,\Omega,t)$ to be

$$\frac{d^2I_\alpha}{d\omega\, d\Omega}=\frac{\sqrt{\kappa}}{4\pi c^3}\langle(\mathbf{e}_\alpha\cdot\hat{\boldsymbol{\mathcal{E}}}^\dagger)_{\mathrm{d}}(-\omega,t)(\mathbf{e}_\alpha^*\cdot\hat{\boldsymbol{\mathcal{E}}})_{\mathrm{d}}(\omega,t)\rangle. \quad (1)$$

Here $(f)_{\mathrm{d}}(\omega,t)=\int_{-\infty}^{\infty}f(t')G(t'-t_{\mathrm{d}}-t)e^{i\omega t'}dt'$, $G(t)$ is a detector function, and t_{d} is a delay time. In terms of the ring's nonequilibrium electric dipole $\mu(t)$, magnetic dipole $\mathbf{M}(t)$, and the quadrupole moment $\mathbf{D}(t)$ (we neglect higher terms) the electric-field coherent part of the emitted light is $\boldsymbol{\mathcal{E}}(\mathbf{R},t)=\hat{\mathbf{n}}\times[\hat{\mathbf{n}}\times\ddot{\boldsymbol{\mu}}(t-t_0)]+\sqrt{\kappa}\{\hat{\mathbf{n}}\times\ddot{\mathbf{M}}(t-t_0)+\frac{1}{6c}\hat{\mathbf{n}}\times[\hat{\mathbf{n}}\times\mathbf{D}(t-t_0)]\}$, with $t_0=R/c$ and unit vector $\hat{\mathbf{n}}\|\mathbf{R}$. We use $G(t)=(\frac{2}{\pi})^{1/4}\frac{1}{\sqrt{\Delta T}}e^{-t^2/\Delta T^2}$, ΔT is the local oscillator-pulse duration [23], and $t_{\mathrm{d}}=t_0$. Below the following polarization vectors are used: $\mathbf{e}_\sigma$ is in the plane of the ring and perpendicular to $\hat{\mathbf{n}}$, $\mathbf{e}_\pi$ is perpendicular to $\mathbf{e}_\sigma$ and $\hat{\mathbf{n}}$, $\mathbf{e}_{\pm45^\circ}=\frac{1}{\sqrt{2}}(\mathbf{e}_\sigma\pm\mathbf{e}_\pi)$, and $\mathbf{e}_\pm=\frac{1}{\sqrt{2}}(\mathbf{e}_\sigma\pm i\mathbf{e}_\pi)$. Our aim is to control the polarization properties of the emitted light by a dynamical control of the rotating charge polarization of the ring, using half-cycle pulses (HCPs) [25,26] or short-laser pulses. Compared to other methods for an ultrafast control of optical fields [27], our emitted radiation is in the subterahertz range and is controllable within times comparable to the charge oscillation period in the ring (cf. below). The light polarization is classified by the Stokes parameters S_0, S_1, S_2, and S_3. S_0 is the intensity, and S_1, S_2, and S_3 quantify, respectively, linearly and circularly polarized light ($P=\sqrt{S_1^2+S_2^2+S_3^2}/S_0$ is the degree of light polarization). In term of (1), S_j read

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$$S_{1(2,3)}(\omega,t;\theta,\phi)=\frac{d^2I_{\sigma(45°,+)}}{d\omega\, d\Omega}-\frac{d^2I_{\pi(-45°,-)}}{d\omega\, d\Omega}. \quad (2)$$

For coherent electric-dipole emission we find if $\hat{\mathbf{n}}$ is perpendicular to the ring, i.e., $\theta=0$ $[S_j^{\perp}\equiv S_j(\theta=0)]$,

$$S_3^{\perp}(\omega,t)=\frac{\sqrt{\kappa}}{4\pi c^3}2\,\mathrm{Im}[(\ddot{\mu}_y)_d(-\omega,t)(\ddot{\mu}_x)_d(\omega,t)]; \quad (3)$$

otherwise, $S_3(\omega,t;\theta,\phi)=S_3^{\perp}(\omega,t)\cos\theta$, whereas $S_0^{\perp}(\omega,t)=\frac{\sqrt{\kappa}}{4\pi c^3}[|(\ddot{\mu}_x)_d(\omega,t)|^2+|(\ddot{\mu}_y)_d(\omega,t)|^2]$, and the ϕ-averaged $S_0(\omega,t,\theta,\phi)$ is given by $S_0^{\perp}(\omega,t)(1+\cos^2\theta)/2$. Further, we introduce $\bar{S}_j(t;\theta,\phi)=2\int_0^{\infty}\frac{d\omega}{2\pi}S_j(\omega,t;\theta,\phi)$, $j=0,1,2,3$. $\bar{S}_0(t,\theta,\phi)$ is a time-dependent power per Ω. The frequency-integrated circular-polarization degree is $P_{\mathrm{circ}}(t;\theta,\phi)=\bar{S}_3(t;\theta,\phi)/\bar{S}_0(t;\theta,\phi)$.

A realization of a "quantum nanosynchrotron" is provided by a spin-degenerate, thin, isolated (without contacts) two-dimensional quantum ring with width d ($d\ll r_0$) containing N carriers. We assume that the ring lies in the xy plane. The single-particle energies ε_{lm} and wave functions $\psi_{lm}(\mathbf{r})$ are labeled by the radial quantum number l and the angular quantum number m. Thereby the angular part of $\psi_{lm}(\mathbf{r})$ is given by $\frac{1}{\sqrt{2\pi}}e^{im\varphi}$ and the radial parts are wave functions of a quantum well with infinite barriers [8]. For clarity we assume d is small enough such that only the lowest radial channel ($l=1$) is populated.[1] In the basis ψ_{lm} the field operator reads $\hat{\Psi}(\mathbf{r},t)=\Sigma_m\hat{a}_m(t)\psi_{1,m}(\mathbf{r})$ and the density matrix is given by $\rho_{mm'}(t)=\langle\hat{a}_m^{\dagger}(t)\hat{a}_{m'}(t)\rangle$ The dipole moment $\mu=\mathrm{Tr}[e\mathbf{r}\rho]$ (e is the electron charge) has the components $\mu_x=2er_0\Sigma_m\mathrm{Re}[\rho_{mm-1}]$ and $\mu_y=2er_0\Sigma_m\,\mathrm{Im}[\rho_{mm-1}]$. The system time scale is given by the ballistic time $\tau_F=2\pi r_0/v_F$, where v_F is the Fermi velocity (typically τ_F is tens of picoseconds). To trigger and control the carrier dynamics, we utilize asymmetric monocycle pulses, so-called HCPs [25,26]. For HCPs the pulse duration is chosen as $\tau_d\ll\tau_F$. Application of the pulse at $t=0$ gives the system a kick, creating a time-dependent dipole moment μ of the ring [5,8]. Theoretically, the parameter α is decisive; $\alpha=r_0p/\hbar$, $p=-e\int_0^{\tau_d}\mathcal{E}_{\mathrm{p}}(t)dt$, and $\mathcal{E}_{\mathrm{p}}(t)$ is the electric field strength of the pulse. For a pulse linearly polarized along the x axis and $\alpha<1$, we find $\mu_x(t)\approx-\alpha er_0\Sigma_m(f_{m-1}^0-f_m^0)\sin[(\varepsilon_m-\varepsilon_{m-1})t/\hbar]$, where $f_m^0=\rho_{mm}(t<0)$ is the equilibrium distribution at $t<0$. Then at $t=\tau_F/4$ we apply a second HCP along the y axis, creating a rotating dipole (and a charge current [7]).

Alternatively, short circular-polarized laser pulses (CPPs) with a frequency centered around $\omega=2\pi/\tau_F$ create a rotating dipole in the ring. Such pulses or their sequences were suggested previously for exciting rotating polarization and currents in atoms [28] and molecular rings [10,17,29], as well as in nanosize quantum rings and dots [9,30]. For a CPP we can define $\alpha'=\frac{1}{2}r_0p'/\hbar$, with $p'=-e\int_0^{\tau_d}\mathcal{S}_{\mathrm{p}}(t)dt$, where $\mathcal{S}_{\mathrm{p}}(t)$ is the envelope of the electric field of the CPP. We have calculated that application of a CPP creates practically the same rotating dipole moment as in the case of the two $\frac{\tau_F}{4}$-delayed perpendicular HCPs with $\alpha=\alpha'$ for not too strong ($\alpha\gg1$) pulses.[2] HCPs are insofar favorable as there is no need to fine-tune the pulse frequency (as for CPPs); also, for disordered or deformed rings it is advantageous to use HCPs. Having created radiating dipoles, it is essential for application to understand how they decay.

Radiative damping results from the back-action of the emitted radiation on the nonequilibrium carriers. Energy loss due to emission causes relaxation of the density matrix. Starting from the light-matter interaction Hamiltonian $\hat{H}_D=-\int d^3r\hat{\Psi}^{\dagger}(\mathbf{r},t)e\mathbf{r}\cdot\hat{\mathcal{E}}(\mathbf{r},t)\hat{\Psi}(\mathbf{r},t)$ the coupled equations for the dynamics of the field and density operators attain a tractable form if we allow for classical (coherent) electric fields only (semiclassical approximation). Applying the adiabatic (Markov) approximation [31,32] we derived the low-temperature decay rate of the dipole moment as $\gamma=\frac{1}{6}\sqrt{\kappa}\frac{e^2\omega_F^2}{m^*c^3}N$, where $\omega_F=2\pi/\tau_F$ is the Fermi angular velocity and m^* is the carrier effective mass. For a semiconductor-based (n-GaAs) ring we have $m^*=0.067m_0$ and $\kappa=12.5$; other typical values are $r_0=1.35$ μm and $N=400$, meaning that $\gamma=1.5\times10^2$ s^{-1}. For $r_0=0.3$ μm and $N=160$ we find $\gamma=0.4\times10^4$ s^{-1}. Considering a planar array of N_r identical rings lying in the xy plane with the array dimension being smaller than the characteristic wavelength of the radiation, the radiative damping rate is found to be $\gamma_{\Sigma}=\frac{1}{6}\sqrt{\kappa}\frac{e^2\omega_F^2}{m^*c^3}NN_r$. In practice, rings have a finite spread Δr_0 of the ring radius distribution ($\Delta r_0\ll r_0$). After a time lapse of $\tau_{\mathrm{spr}}=\frac{2\pi}{\omega_F}\frac{r_0}{\Delta r_0}$ the electron dynamics in different rings is out of phase and the intensity of coherent radiation vanishes. Hence, the value of γ_{Σ} is relevant only at short times after the array excitation; otherwise, it provides an upper bound.

To account for the spontaneous emission contribution to the radiative damping we study the correlations between the field and density operators [32] by applying the Hartree-Fock approximation $\langle\hat{a}_{m_1}^{\dagger}\hat{a}_{m_2}^{\dagger}\hat{a}_{m_3}\hat{a}_{m_4}\rangle=\rho_{m_1m_4}\rho_{m_2m_3}-\rho_{m_1m_3}\rho_{m_2m_4}$ and neglecting correlations involving two photon operators. The adiabatic approximation for the field-density correlations leads to additional contributions to the decay of the nondiagonal density matrix components $\rho_{mm'}$ due to the spontaneous emission; namely, $\gamma_{mm'}^{\mathrm{sp}}=\gamma(\frac{\varepsilon_m-\varepsilon_{m'}}{\hbar\omega_F})^3(2+f_{m+1}^0+f_{m'+1}^0-f_{m-1}^0-f_{m'-1}^0)$. Note that the effective spontaneous decoherence rate (decay rate of the dipole moment) does not exceed 2γ for low temperatures.

For weak excitation ($\alpha<1$) optical phonons are not involved due to their high energy. Relaxation of the excited population is, however, caused by scattering from longitudinal acoustic phonons (the corresponding electron-phonon interaction Hamiltonian is described, e.g., in Ref. [8] and is similar to the light-matter interaction Hamiltonian). Proceeding as for the radiative damping and assuming the Debye model for the phonon dispersion, we derived the decay rate of the dipole moment due to the emission of *coherent pho-*

[1]Theory is extendable to multiple radial channels as in [6].

[2]Note that the number of cycles of a CPP is not too large (its frequency distribution is not too narrow) so that all transitions between neighboring levels of the ring in the neighborhood of the Fermi level can take place.

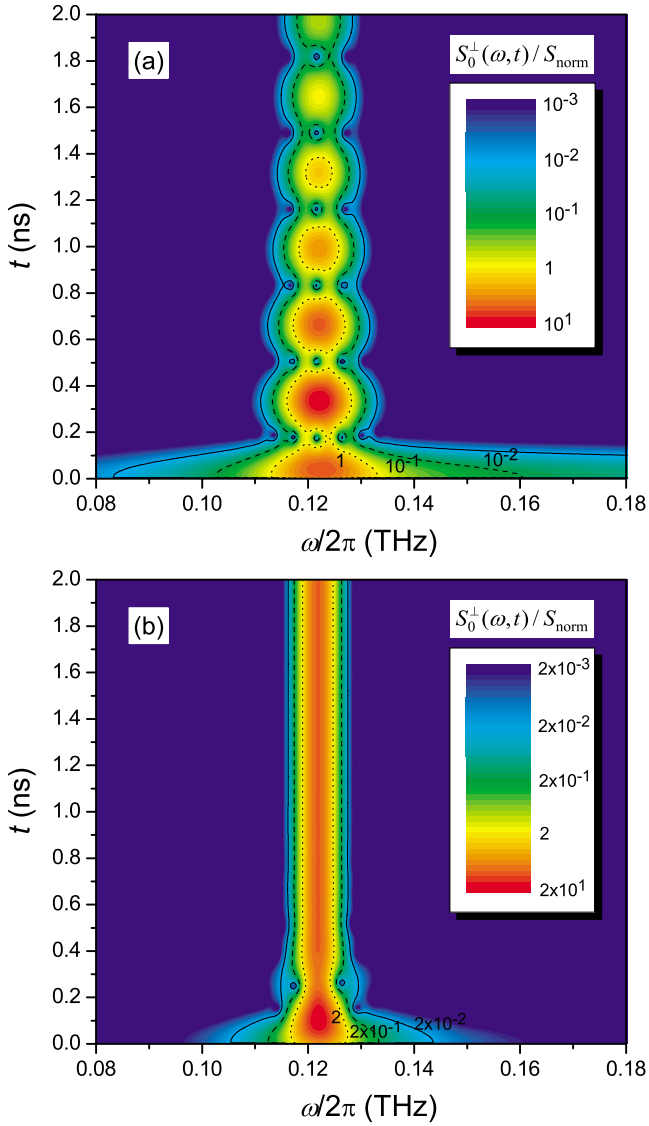

FIG. 1. (Color online) Time-dependent spectrum of the radiation emitted from a quantum ring. The spectrum is normalized to $S_{\rm norm}=\sqrt{\kappa}\frac{e^2r_0^2\omega_F^3}{4\pi c^3}$. Parameters of the quantum ring are $r_0=0.3$ μm, $d=20$ nm, and $N=160$. The ring is excited (a) by two mutually perpendicular HCPs with kick strength $\alpha=0.4$ and delay time $\tau_F/4$ at $T=4$ K and (b) by a periodic sequence of two mutually perpendicular HCPs with kick strength $\alpha=0.1$, delay time $\tau_F/4$, and period τ_F, at $T=10$ K. The duration of each HCP is $\tau_d=0.5$ ps, peak field value 33.6 V/cm in (a) and 8.4 V/cm in (b). Detector time is $\Delta T=100$ ps.

non waves as $\gamma^s=\frac{1}{\tau_{\rm LA}}F(\frac{\omega_F d}{c_{\rm LA}})$ if $\omega_F<\omega_{\rm D}$, a condition valid for our rings. Here $\omega_{\rm D}$ is the Debye frequency, $c_{\rm LA}$ is the LA velocity of sound, the function $F(y)$ for our quantum-well radial confinement is $F(y)=8\pi^2 y\int_0^y dx\frac{1}{\sqrt{1-x^2/y^2}}\frac{\sin^2(x/2)}{x^2[x^2-(2\pi)^2]^2}$, and $\frac{1}{\tau_{\rm LA}}=\frac{|D|^2}{\hbar c_{\rm LA}^2\rho_s d^2 r_0}$, where D is the deformation constant and ρ_s is the lattice density. For electrons in GaAs we find $\gamma^s=0.8\times10^6$ s^{-1} for the case $r_0=1.35$ μm, $d=50$ nm, and $N=400$. For $r_0=0.3$ μm, $d=20$ nm, and $N=160$ we deduce $\gamma^s=2.1\times10^8$ s^{-1}. Note that the wavelength of emitted phonons is less or on the order of the ring size. Considering thus a planar ring array the phonon waves emitted from different places are out of phase; a large increase in the damping constant with the number of rings is thus unlikely. Density matrix relaxation due to scattering by *incoherent phonons* we considered in Ref. [8], based on which we conclude that the nondiagonal density matrix components decay due to the spontaneous emission of incoherent phonons by the rate $\gamma_{mm'}^{s,\rm sp}=\frac{1}{\tau_{\rm LA}}\Sigma_\nu(R_m^{m+\nu}+R_{m'}^{m'+\nu})$, where $R_{m'}^m=F(q_{m'}^m d)\chi_{m'}^m f_m^0$ if $q_{m'}^m\in(0,\frac{\omega_{\rm D}}{c_{\rm LA}})$, $R_{m'}^m=F(q_{m'}^m d)\chi_{m'}^m(1-f_m^0)$ if $q_{m'}^m\in(-\frac{\omega_{\rm D}}{c_{\rm LA}},0)$, and $R_{m'}^m=0$ otherwise. Here $q_{m'}^m=(\varepsilon_m-\varepsilon_{m'})/(\hbar c_{\rm LA})$, $\chi_{m'}^m=1$ if ${\rm sgn}(m)={\rm sgn}(m')$ and $|q_{m'}^m|<\frac{\omega_{\rm D}}{c_{\rm LA}}$, $\chi_{m'}^m=0$ otherwise. Using the expression for $\gamma_{mm'}^{s,\rm sp}$, we calculated numerically the effective decay rate of the dipole moment for different parameters of the quantum ring. For the above-mentioned ring parameters the decay rate rises from 10^8 s^{-1} at $T=1$ K to 10^{10} s^{-1} at $T=10$ K (see also Ref. [8]). Thus, scattering by incoherent phonons is the main source for our dipole moment relaxation.

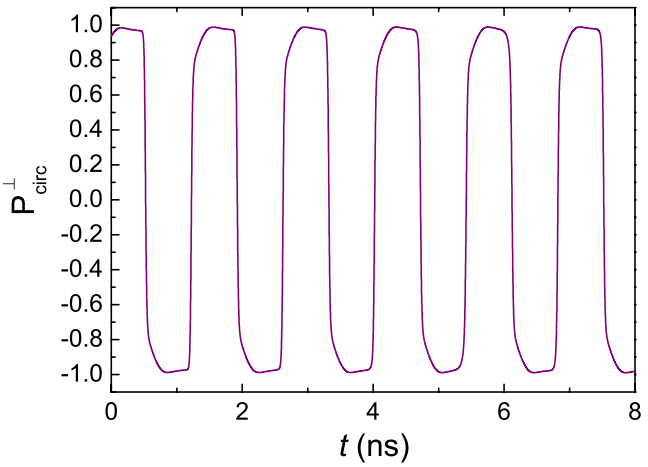

FIG. 2. (Color online) Dynamics of the detected circular polarization. The ring parameters are $r_0=1.35$ μm, $d=50$ nm, $N=400$, and $T=4$ K. For the parameters of the excitation, see the text. Detector time is $\Delta T=100$ ps.

For illustration we focus on a quantum ring excited by two mutually perpendicular HCPs delayed by $\tau_F/4$. Figure 1 shows the time-dependent spectrum of the radiation emitted perpendicular to the ring plane ($\theta=0$). Only the electric dipole contributes to the radiation spectrum in this geometry. Calculating the time-dependent spectrum we use the detector time $\Delta T=100$ ps. The numerically calculated time-dependent spectrum in Fig. 1 exhibits repeated light bursts centered at approximately ω_F and with decaying peak values due to the relaxation pathways discussed above. The bursts correspond to revivals of the charge polarization dynamics [8]. The abrupt radiation switch-on upon charge-polarization generation results in a relatively broad spectrum at short times. Then we consider the degree of circular polarization at $\theta=0$, which we denote as $P_{\rm circ}^{\perp}(t)$. For a detected frequency range $[0.5\omega_F,1.5\omega_F]$ we find $P_{\rm circ}^{\perp}(t)>0.99$. Driving the rotating dipole in sequence effectively stabilizes the time-dependent spectrum, reaching a practically stationary state [cf. Fig. 1(b)]. In this case $P_{\rm circ}^{\perp}(t)>0.999$ is achievable for the mentioned frequency range.

Having demonstrated the possibility of an ultrafast generation of circular-polarized subterahertz light, we are interested in the ultrafast control of the degree of the circular polarization. For illustrating the ubiquitous nature of the effect, we choose a different radius of 1.35 μm. The pulses parameters are $\alpha=0.2$, $\tau_d=3$ ps, and a time delay $\tau_F/4$. Furthermore, for a periodic train of two mutually perpendicular HCPs, we assume a period of 0.7 ns. Each applied pulse sequence changes the sense of rotation of the ring dipole moment. This leads to a sign change of $P^{\perp}_{\mathrm{circ}}$, as demonstrated in Fig. 2; i.e., the photons are emitted in portions with an alternating helicity. Generally, more complicated pulse sequences allow control of the chirality of each emitted photon portion.

Summarizing, we developed a theory for the ultrafast, time-dependent detection of the light polarization properties and applied it to the emission from quantum rings. As an illustration, we presented calculations for the time-dependent spectrum, accounting for relaxation effects. Applying appropriate sequences of half-cycle pulses (or short circular polarized laser pulses) allows one to generate circular-polarized subterahertz light on picosecond time scales and to change and control the degree of the circular polarization of the emitted photons in subnanoseconds. For a sufficient intensity of emitted light, one can either use an array of rings [11,16,33] or excite a single ring far beyond the weak excitation regime considered in the present paper. Our treatment is directly applicable to the first case whereby a small dispersion of the ring sizes in the array must be provided. The proposed theory is extendable to higher excitations and to quantum dots and quantum rings with an impurity [34] as well as to graphene rings [35] and chiral and nonchiral molecular rings [10,17,29,36,37] and to atoms [28]. Finally, the developed theory is applicable to the time-dependent detection of the circular polarization of quantum light [38].

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