A predicted “Faraday oscillation” in photoexcited semiconductors

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While, for semiconductors photoexcited by a circularly polarized pump, the polarization plane of a linearly polarized probe has been shown to rotate, we here predict a spectacular change when the pump beam is linearly polarized, from Faraday rotation to “Faraday oscillation”, the oscillation of the polarization plane going along a change of the photon polarization from linear to elliptical.

This effect, which reduces to zero when the probe field is parallel or perpendicular to the pump field, comes from coherence between the real excitons created by the pump and the virtual exciton coupled to the unabsorbed probe — as easy to see from the Shiva diagrams which represent the many-body physics taking place in this coupled photon-composite-exciton system.

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Faraday rotation is a physical effect known for quite a long time [1]. It takes place in “optically active media” which have different refractive indices $n_{\pm}$ for photons with circular polarization: the two parts of a linearly polarized probe beam then travel at different speeds, making the polarization plane of this probe beam rotate around the propagation axis, with a rotation proportional to $(n_+ - n_-)$. The refractive index difference physically comes from a dissymmetry of the material which can preexist or be induced. The standard way to induce this dissymmetry is through a magnetic field which produces an energy difference between electrons with up and down spins. In semiconductors, this dissymmetry can also be produced by the absorption of a circularly polarized pump beam [2-13]. In a recent work [14], we have shown, by using the many-body theory we have developed for composite bosons [15], that the real excitons created by the pump interact with the virtual excitons coupled to the $\sigma_\pm$ parts of the unabsorbed probe, not only through Coulomb interaction, but mainly through the Pauli exclusion principle between the fermionic components of these excitons. With a $\sigma_+$ pump acting on quantum well, this exclusion — which is directly linked to the exciton composite nature — only exists with the virtual excitons coupled to the $\sigma_+$ part of the linearly polarized probe. The resulting difference in the response function $S_{\pm}$ to $\sigma_{\pm}$ photons leads to $n_+ \neq n_-$. We here consider a far more subtle situation: If instead of using $\sigma_+$ photons, we create excitons through the absorption of a pump beam having a linear polarization along $x$, a naïve thinking would lead to say that the $\sigma_+$ and $\sigma_-$ parts of a linearly polarized probe see the $x$ excitons — which are half $\sigma_+$ and half $\sigma_-$ — in the same way, so that the refractive indices $n_{\pm}$ should be the same. This naïve thinking however forgets the importance of coherence between the $\sigma_{\pm}$ parts of the pump and the probe: It is somewhat obvious that, in the same way as excitons created by a $\sigma_+$ pump beam act differently on the $(\sigma_+, \sigma_-)$ parts of a linearly polarized probe, excitons created by a $x$ pump must affect the $(x, y)$ parts of the light differently.

In this letter, we predict a spectacular change when the polarization of the pump goes from circular to linear. Indeed, a rotation of the polarization plane implies to break the clockwise-counterclockwise invariance of the system. This happens with excitons created by $\sigma_+$ photons, but not by a linearly polarized pump. With such a pump, the probe polarization plane has no reason to

FIG. 1: (a) Time evolution of the probe photon ellipticity when, for $t = 0$, the probe has a linear polarization along $\theta_0$ with $0 < \theta_0 < \pi/4$, while the pump excitons are polarized along $x$. The ellipticity change goes along an oscillation of the polarization main axis around $x$. (b) Same as (a) for $\pi/4 < \theta_0 < \pi/2$, the oscillation taking place around $y$. (c) When $\theta_0 = \pi/4$, the probe photon polarization goes from linear to circular, its main axis staying at $\pi/4$ from $(x, y)$. It then returns to linear but with a main axis at $(-\pi/4)$. And so on...
rotate one way or the other; if something happens, this cannot be a rotation. We here show that, in the case of a $x$ pump, an unabsorbed probe beam polarized along $x$ or $y$ stays unchanged, while for any other direction $\theta_0 \neq (0, \pi/2)$, the polarization changes from linear to elliptical, the ellipse main axis oscillating around $x$ or $y$:

When $0 < \theta_0 < \pi/4$, the ellipticity of the probe photon first increases while the main axis rotates from $\theta_0$ to $0$. This ellipticity then decreases while the main axis keeps rotating from $0$ to $-\theta_0$ where it turns linear again (see Fig.1(a)). For $\pi/4 < \theta_0 < \pi/2$, a similar oscillation takes place around $y$ (see Fig.1(b)). Continuity between these two oscillations is obtained for $\theta_0 = \pi/4$ with a probe beam which goes from linear to circular, its main axis staying along $\theta_0 = \pi/4$. The probe photon ellipticity then shrinks to become linear again, but with a main axis along $(-\pi/4)$, where it returns to circular, the main axis staying along $(-\pi/4)$, and so on... (see Fig.1(c)).

**Physical idea.** This behavior follows from the idea that eigenstates for photons coupled to excitons linearly polarized along $x$, have an $x$ or $y$ polarization. The time evolution of a linear polarization along $\theta_0$ for $t = 0$ then reads

$$\mathbf{e}_t = \mathbf{x} e^{-i\Omega e t} \cos \theta_0 + y e^{-i\Omega e t} \sin \theta_0 .$$

By writing this polarization as

$$\mathbf{e}_t = e^{i\varphi_t} (\mathbf{X}_t \cos \xi_t + i \mathbf{Y}_t \sin \xi_t) ,$$

which describes photons with ellipticity $\xi_t$ and main axis ($\mathbf{X}_t, \mathbf{Y}_t$), we find from ($\mathbf{e}_t, \mathbf{X}_t$) and ($\mathbf{e}_t, \mathbf{Y}_t$),

$$e^{2i\xi_t} = \cos 2\theta_0 \cos 2\alpha_t + \sin 2\theta_0 (\sin 2\alpha_t \cos \Delta_t + i \sin \Delta_t) ,$$

where $\Delta_t = (\Omega_x - \Omega_y)t$ while $\alpha_t$ is such that $\mathbf{X}_t = x \cos \alpha_t + y \sin \alpha_t$.

Equation (3) readily gives the time evolution of the ellipticity as

$$\sin 2\xi_t = \sin 2\theta_0 \sin \Delta_t .$$

This shows that photons stay linearly polarized for $\theta_0 = (0, \pi/2)$, i.e., when the polarization is along $x$ or $y$, while for $\theta_0 = \pi/4$, the photon ellipticity increases up to circular, which is reached for $\Delta_t = (\pi/2, 3\pi/2, \cdots)$.

From the modulus of eq. (3), we find the time evolution of the photon polarization main axis as

$$\tan 2\alpha_t = \tan 2\theta_0 \cos \Delta_t .$$

This shows that $\mathbf{X}_t$ oscillates from $\alpha_t = \theta_0$ to $\alpha_t = -\theta_0$ when $0 < \theta_0 < \pi/4$, while for $\pi/4 < \theta_0 < \pi/2$, it oscillates from $\theta_0$ to $(\pi - \theta_0)$ (in order to stay unchanged for $\theta_0 = \pi/2$). The intermediate value $\theta_0 = \pi/4$ can appear as singular, since $\alpha_t$ jumps from $\pi/4$ to $-\pi/4$ when $\Delta_t$ passes $\pi/2$. However, for $\Delta_t = \pi/2$, the photon polarization is then circular, so that the “main axis” $\pi/4$ and $-\pi/4$ are totally equivalent. Equations (4,5) thus support the time evolution of the probe polarization shown in Fig.1.

Before going further, let us mention that the standard Faraday rotation, obtained with circularly polarized pump photons, can be recovered along the same line. Indeed, the $t = 0$ probe photon polarization given in eq. (1), reads in terms of the $\sigma_{\pm}$ polarization $\mathbf{e}_{\pm} = (x \pm iy)/\sqrt{2}$ as

$$\mathbf{e}_{t=0} = (e^{-i\theta_0} \mathbf{e}_+ + e^{i\theta_0} \mathbf{e}_-)/\sqrt{2} .$$

When the photon eigenstates have a circular polarization — as obtained with excitons created by a $\sigma_+$ pump — the probe photon polarization evolves according to

$$\mathbf{e}_t = \left[ e^{-i\theta_0} \mathbf{e}_+ e^{-i\Omega e t} + e^{i\theta_0} \mathbf{e}_- e^{-i\Omega e t} \right] / \sqrt{2} .$$

Within an irrelevant phase factor $\varphi_t = -(\Omega_+ + \Omega_-)t/2$, this corresponds to probe photons staying linearly polarized with a polarization plane rotating as $\theta_t = \theta_0 + (\Omega_+ - \Omega_-)t/2$.

**Photon eigenstates.** Let us now show the link between the photon eigenstates and the pump polarization $\mathbf{e}_p$. As $\mathbf{e}_p = x \cos \xi_p + iy \sin \xi_p$ also reads $\mathbf{e}_+ \cos \xi_p' + \mathbf{e}_- \sin \xi_p'$, with $\xi_p' = \pi/4 - \xi_p$, this pump gives rise, if we neglect spin relaxation, to $N$ coherent excitons $B^\dagger_p = \cos \xi_p'B_{o+1} + \sin \xi_p'B_{o-1}$, where $B^\dagger_p$ creates a ground state exciton with spin $S$. An unabsorbed probe photon with polarization $S = \pm 1$ interacts with these pump excitons through the virtual excitons $B^\dagger_p$ to which they are coupled. The lowest order term in the pump exciton density comes from processes in which one virtual exciton interacts with one among $N$ pump exciton. As shown in our previous work on Faraday rotation [14], these processes are represented by the four Shiva diagrams [16] of Fig.2, in which the excitons coupled to the “in” and “out” probe photons have 2, 1 or 0 common carriers — with possibly some additional Coulomb processes.

Due to spin conservation when the “in” and “out” excitons are made from the same pairs, we readily see that the diagram (2a) has a polarization prefactor $\partial_{S,S}(\cos^2 \xi_p' + \sin^2 \xi_p')$, while the prefactor for the diagram

![](FIG.2: Possible processes between $\sigma_S$ photons (wavy lines) coupled to ($S = \pm 1$) virtual excitons, these composite excitons interacting with one pump exciton $P$ having a polarization $\cos \xi_p' (+1) + \sin \xi_p' (-1)$ by Coulomb interaction (diagrams a,d) or just by carrier exchange (diagrams b,c).)


we multiply it by $P^{|\sigma_S^{(1)}\rangle\langle\sigma_S^{(1)}|}$. The diagrams (2b,2c), in which the excitons exchange one carrier, we must remember that, as, in quantum wells, carrier exchanges between $(+1,-1)$ excitons lead to dark states $(\pm 2)$, exchanges can only exist between the $(+1)$ parts or the $(-1)$ parts of these excitons [17]. This leads to the same polarization prefactor $\delta_{S',S}(\delta_{S,+1}\cos^{2}\xi_p' + \delta_{S,-1}\sin^{2}\xi_p')$ for the diagrams (2b,2c).

By calling $(D_a, D_d)$ the orbital parts of the direct diagrams (2a,2d) and $(X_b, X_c)$ the orbital parts of the exchange diagrams (2b,2c), the eigenstates for probe photons coupled to pump excitons $P = \cos \xi_p^{(1)}(+1) + \sin \xi_p^{(1)}(-1)$, are obtained in the $(+1,-1)$ basis, from

$$
\begin{vmatrix}
D_a + X\cos^{2}\xi_p' - \Omega & D_d\sin\xi_p'\cos\xi_p' \\
D_d\sin\xi_p'\cos\xi_p' & D_a + X\sin^{2}\xi_p' - \Omega
\end{vmatrix} = 0, \quad (9)
$$

where $X = X_b + X_c + D_d$, the only contribution not in $\delta_{S',S}$ coming from diagram (2d) through eq. (8). Equation (9) shows that (i) for a $\sigma_+\uparrow$ pump, $\xi_p = \pi/4$, i.e., $\xi_p' = 0$, the photon eigenstates are $S = \pm 1$: They have a circular polarization, with an energy splitting $\Omega_+ - \Omega_- = X$. (ii) For a pump linearly polarized along $x$, $\xi_p = 0$, i.e., $\xi_p' = \pi/4$, the eigenstates $[(+1) \pm (-1)]\sqrt{2}$ have linear polarizations along $x$ and $y$, the energy splitting being $\Omega_+ - \Omega_y = D_d$.

Consequently, the physical processes responsible for the Faraday rotation induced by a $\sigma_+\uparrow$ pump, which makes $\Omega_+ \neq \Omega_-$, are the ones in which the pump exciton and the virtual exciton coupled to the probe exchange 1 or 2 carriers (see Figs.2(b,c,d)), as previously found using another procedure [14]. On the opposite, the Faraday oscillation produced by an $x$ pump which makes $\Omega_x \neq \Omega_y$, only comes from the process of Fig.2(d) in which the two excitons exchange their two carriers.

**Formalism.** Let us now outline the formalism which leads to eq. (9) for the eigenstates of a probe photon in the presence of $N$ coherent excitons $B_P^{1\dagger}$. In the absence of semiconductor-photon coupling, the two states $|\phi_S\rangle = a_P^{1\dagger}|v\rangle \otimes B_P^{1\dagger}|v\rangle$, where $a_P^{1\dagger}$ creates a photon with polarization $S = \pm 1$, are degenerate in energy, $(H_0 - E_0)|\phi_S\rangle = 0$. The eigenstates of the coupled system $(H - \mathcal{E})|\psi\rangle = 0$, with $H = H_0 + W, W = U + U^\dagger$ and $U = \sum_j \mu_j B_j^{1\dagger}\alpha_S$, are easy to obtain by writing $|\psi\rangle$ as

$$
|\psi\rangle = \langle \tilde{\phi}_{+1}|\tilde{\phi}_{-1}\rangle + P_{\uparrow}\langle \tilde{\phi}_{+1}|\tilde{\phi}_{-1}\rangle + P_{\downarrow}|\psi\rangle, \quad (10)
$$

where $|\tilde{\phi}_S\rangle$ is the normalized state $|\phi_S\rangle$, while $P_{\uparrow}$ is the projector over the subspace perpendicular to $|\tilde{\phi}_{\pm1}\rangle$. If we insert this $|\psi\rangle$ into the Schrödinger equation and we multiply it by $P_{\uparrow}$, we get, to lowest order in the semiconductor-photon coupling,

$$
P_{\uparrow}|\psi\rangle \simeq P_{\uparrow}\frac{1}{E_0 - H_0} P_{\downarrow} W \sum_S |\phi_S\rangle \langle \phi_S|\phi_S\rangle |\psi\rangle. \quad (11)
$$

The state $U^\dagger|\phi_S\rangle$ contains $N$ pump excitons and one virtual exciton with spin $S$ coupled to a $S$ photon. The contribution to $\Delta_{S',S}$ linear in the exciton density, corresponds to the interactions of this virtual exciton with one among $N$ pump excitons [14]. For photon detuning close enough to the exciton ground state, in order to only keep the photon coupling to this exciton, $\Delta_{S',S}$ reduces to

$$
\Delta_{S',S}^{(1)} = N|\mu_S|^2\langle v|B^\dagger F B_{0S'} \frac{1}{\delta + 2E_0 - H_{sc}} B^\dagger_{0S} B^\dagger_{F} |v\rangle, \quad (14)
$$

where $E_0$ is the exciton ground state energy, $\delta$ the photon detuning and $H_{sc}$ the semiconductor Hamiltonian. $\Delta_{S',S}^{(1)}$ corresponds to the diagram of Fig.3(a). Since the two-pair-eigenstate spectrum is not known, $\Delta_{S',S}^{(1)}$ cannot
be calculated exactly for any detuning. Eq.(14) already shows that the smaller the detuning, the larger the coupling. We can also say that, as carrier exchanges between excitons (+1) and (−1) lead to dark states [17], the non-diagonal coupling $\Delta_{1,-1}^{(1)}$ responsible for the Faraday oscillation can only come from direct Coulomb processes in which the excitons $B_{o,1}$ and $B_{o,-1}$ are made with different electron-hole pairs, like in diagram (2d). However as, for exciton momenta close to zero, the direct Coulomb scattering $\xi (\alpha \sigma)$, which corresponds to the first diagram of Fig.3(b), reduces to zero [18], $\Delta_{1,-1}^{(1)}$ at large detuning is controlled by direct processes in which enter two Coulomb interactions at least (see Fig.3(b)). Dimensional arguments then lead to a non-diagonal coupling $\Delta_{1,-1}^{(1)}$ in $(R_X/\delta)^2$ compared to the diagonal coupling $\Delta_0^{(1)}$ controlled by exchange processes in the absence of Coulomb interaction, i.e., diagrams (2b,2c). Consequently, at large detuning, the energy splitting producing the Faraday oscillation is much smaller than the one responsible for the Faraday rotation: To observe this new Faraday oscillation, we thus need very good samples in order to have a narrow exciton line to possibly approach the exciton resonance without sizeable residual absorption.

Experiments on Faraday rotation or Kerr effect in photoexcited semiconductors have been reported by various groups [2,3,5,13,19,20]. In these experiments, the probe photons are at resonance, while for a “pure” effect, we should use unabsorbed photons in order to be coupled to virtual excitons. These experiments however show that, with a $x$ pump, photons polarized along $x$ or $y$ stay unchanged [19,20], in agreement with the present work.

Note that, while Faraday rotation here appears through the time evolution of the probe polarization in the presence of pump excitons, this effect is usually measured when passing through a photoexcited sample. The two points of view can be related through the time $t$ the light travels in the sample. Also note that the present approach allows to point out the role of coherence between pump and probe excitons, in contrast to standard approaches in which the response functions to $\sigma_+$ and $\sigma_-$ photons are calculated separately.

Conclusion. The many-body theory for composite bosons we have recently constructed [15] is quite appropriate to predict subtle polarization effects like the ones induced by exciton coherence. While the precise understanding of the last part of this letter requires some background on this theory — which can be found in various previous works [15] — the spectacular change from Faraday rotation to Faraday oscillation just follows from accepting the physically reasonable idea that, in the same way as the eigenstates for photons coupled to $\sigma_+$ excitons have a $(\sigma_+, \sigma_-)$ polarization, the ones coupled to $x$ excitons have a $(x,y)$ polarization, this change being rather obvious from the Shiva diagrams [16] which represent the interactions of a pump exciton and a virtual exciton coupled to the probe.

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