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Nonlinear Optical Gyrotropy

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We discuss some general aspects regarding the nonlinear optical gyrotropy with particular emphasis on the photoinduced modifications and control of the polarization state of a beam through light self-action. Both artificial and natural gyrotropic materials are considered and we show that in the former class the magnetooptic interaction can lead to a striking behavior because of the nonreciprocity. We discuss different configurations of nonlinear interactions in gyrotropic media with certain applications and estimations of the relevant coefficients.

Keywords: Self-action; gyrotropy; magnetooptic effect

INTRODUCTION

The polarization state of the light reflects [1, 2] the vector nature of the electromagnetic field, in particular the interplay between its electric and magnetic parts, and introduces topological features that have important implications regarding discrimination and robustness of certain electromagnetic interactions. This has already found numerous and important optical applications in the linear regime [3] but the nonlinear regime offers in addition the possibility to modify and control the polarization state of a beam through light self-action or photoinduced processes with far-reaching repercussions in applications as well. This can be particularly striking when gyrotropy, or the ability of a material to rotate the polarization state, and nonreciprocity come

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into play as in the case of magnetooptical interactions; gyrotropy is also encountered in optical activity, a manifestation of nonlocality in the field-matter interaction.

Notwithstanding the fundamental interest and technological potential of such aspects their study has not been pursued to any consequence in nonlinear optics. One reason is the complications engendered by the nonlocal character of electromagnetic interactions and the intermingling of electric and magnetic effects that in essence underlie gyrotropy and contrast with the commonly made assumption in nonlinear optics [4, 5] that the matter-field interaction is local and only electric effects are involved (electric dipole approximation). Another important reason is the apparent smallness of the physical parameters that govern such nonlocal effects unless certain provisions are made to enhance them with concommitant complications. However both reasons loose much of their deterring impact when the relevant effects are put in the proper perspective; the gyrotropic interactions indeed address features, namely polarization state rotations, that allow outstanding discrimination [3, 6] against the background of those encountered in the usual nonlinear optical processes. In addition the gyrotropic nonlinear optical interactions are of much fundamental interest *per se* and their exploitation opens some intriguing and novel perspectives.

Here we summarily discuss some aspects related to photoinduced gyrotropy [7], namely the case where the gyrotropic power of a material can be modified by the light itself, and some of its implications in nonlinear optical propagation. Particular attention will be given in configurations where non-reciprocity is also present for instance in the case of artificial gyrotopy due to the presence of a static magnetic field and can lead to interesting technological developments. This is, for instance, the case when unidirectional control or shielding of optical signal transfer is an issue, or in connection with storage and transfer of coherence, quantum optical or spin coherence in particular.

GYROTROPY. GENERAL ASPECTS

Optical gyrotropy or circular birefringence and circular dichroism is the ability of a material to rotate [1–6] the plane of polarization of an

incident linearly polarized plane wave after propagation inside the material. All materials may naturally or artificially possess this property. Irrespective of the physical mechanisms that underlie this property, the constitutive relation between an oscillating electric field $\underline{E}$ and electric induction $\underline{D} = \underline{E} + 4\pi\underline{P}$ in a gyrotropic medium where $\underline{P}$ is the induced polarization can be cast [1, 2] in the form

$$\underline{D} = \underline{\underline{\varepsilon}}\,\underline{E} + i(\underline{E} \wedge \underline{g}) \tag{1}$$

Here, $\underline{g}$ is the gyration vector, which for isotropic media and crystals with cubic symmetry is parallel to the wavevector $\underline{k}$ in the natural gyrotropy (optical activity/natural rotatory power) and to the externally applied static magnetic field $\underline{H}_0 = H_0\hat{z}$ in the artificial one (Faraday rotation); in anisotropic bodies, the relation between $\underline{g}$ and $\underline{k}$ or $\underline{H}_0$ is tensorial. We shall disregard effects of higher than first order in $\underline{k}$ or $\underline{H}_0$; here, $\underline{k} = (\omega n/c)\hat{k}$ where c is the light velocity in vacuum and n is the refractive index at frequency ω.

The most striking implication of the vector product in Eq. (1) is that it lifts the degeneracy between left $(-)$ and right $(+)$ circularly polarized modes inherent in all bodies when $\underline{g} = 0$. This is the circular birefringence/dichroism, namely that the refractive/absorptive indices are different for left and right circularly polarized modes with the two modes now propagating with different phase velocities, c/n_- and c/n_+, respectively. This can be readily deduced from the dispersion relation

$$n^2[\hat{e} - \hat{k}(\hat{k} \cdot \hat{e})] - n_0^2\hat{e} + i(\hat{e} \wedge \underline{g}) = 0 \tag{2}$$

which is simply obtained [1–3] by inserting relation (1) in the propagation equation

$$\underline{\nabla} \wedge (\underline{\nabla} \wedge \underline{E}) = -\ddot{\underline{D}}/c^2 \tag{3}$$

in the linear regime. Neglecting terms higher than linear in $\underline{g}$ relation (2) can also be written

$$\left(\underline{k} \pm \frac{1}{2n_0}\underline{g}\right)^2 = n_0^2 \tag{4}$$

or $n_\pm^2 = n_0^2 \pm g_z$ for the left and right refractive indices with $n_0^2 = \varepsilon_0$, the linear dielectric constant. The polarization state of a linearly

polarized beam undergoes a rotation by an angle

$$\theta_R = \frac{\omega L}{2c}(n_- - n_+) \tag{5}$$

after propagation over a distance L inside the medium. The polarization eigenmodes are now the left and right circularly polarized ones $E_\pm = e^*_\pm \cdot \underline{E}$ with $\hat{e}_\pm = (\hat{e}_x \pm i\hat{e}_y)/\sqrt{2}$.

Although $\underline{g}$ is along $\underline{k}$ in both the artificial and natural gyrotropy, there is an essential difference [1–3] between the two classes most easily grasped in the case of isotropic or cubic medium with $\underline{g} = f\hat{k}$, where f is a material property and $\underline{k}$ is along the z-axis. For a naturally gyrotropic medium one obtains from (2) $E_x = \pm iE_y$ irrespective of the direction of the wavevector while for an artificially gyrotropic medium (with a magnetic field present along the z-axis) one gets $E_x = \pm iE_y$ for one propagation direction and the opposite sign for the reversed one with respect to the magnetic field. This is the nonreciprocity previously referred to and is a consequence of the breakdown of the time-reversal symmetry that a static magnetic field introduces and can also be viewed as an artificial irreversibility without the presence of a "bath". The distinction between natural and artificial gyrotropy is most striking when the beam is normally reflected back into the gyrotropic medium, and retraces its path in the backward direction: at each point, its rotation angle is restored in a natural gyrotropic medium while it doubles in an artificially one; the Faraday effect is thus nonreciprocal. More generally the nonreciprocity is manifested whenever a property is non invariant under the transformation $E_\pm \rightarrow E^*_\mp$.

In an intense light beam, the optical properties of a medium undergo [4, 5] modifications and become field amplitude dependent. Of particular interest here are the photoinduced modifications that are proportional to components of the bilinear form $\underline{\underline{P}} = (cn/2\pi)\underline{E} : \underline{E}^*$ and do not involve any frequency shifts; we remind that the light intensity (magnitude of the Poynting vector) is $I = \mathrm{Tr}\,\underline{\underline{P}}$. One such photoinduced modification affects the dielectric constant which becomes $\tilde{\varepsilon} = \varepsilon_0 + \varepsilon_2 I$ or $\tilde{n} = n_0 + n_2 I$ in the simplest scalar case and this is the extensively studied optical Kerr effect that among other nonlinear optical processes underlies the photoinduced linear birefringence, optical phase conjugation, optical bistable operation in a resonator and soliton formation in a fiber.

Similarly, one can have photoinduced gyrotropy [7] or photoinduced circular birefringence/dichroism connected with photoinduced modification of the gyration vector or equivalently $\tilde{f}=f_0+f_2 I$ in an isotropic medium, keeping in mind the previously evoked distinction between natural and artificial gyrotropy. Referring to (4) this results in photoinduced modifications by different amounts of the left and right indices or $\tilde{n}_+ = n_+ + n_{2+} I$ and $\tilde{n}_- = n_- + n_{2-} I$ and accordingly the polarization rotation angle for a linearly polarized incident beam now becomes

$$\tilde{\theta}_R = \frac{\omega L}{2c}(\tilde{n}_- - \tilde{n}_+) = \frac{\omega L}{2c}(\Delta n + \Delta n_2 I) = \theta_R + \theta_2 I = \theta_R + \theta_{NL} \qquad (6)$$

where $\Delta n = n_- - n_+$, $\Delta n_2 = n_{2-} - n_{2+}$ and θ_R is given by (5); $\theta_{NL} = \theta_2 I_{NL} = \theta_2 I$ is the light (photo) induced modification of the rotation angle.

The previous considerations with the inherent simplifications related to the assumed isotropy of the medium can be recast in a rigorous and general form by consistently introducing the relevant nonlinear polarization sources expressed in terms of the phenomenological (macroscopic) nonlinear susceptibilities and the appropriate combinations of field amplitudes; expressions of these polarization sources and the corresponding susceptibilities can be derived [4, 5] from microscopic quantum mechanical considerations and modelling of the matter-field interaction.

Thus in the case of natural gyrotropic media the polarization up to cubic terms in E can be written

$$\begin{aligned} \underline{P} = {} & \underline{\underline{\chi}}^{(1)} \underline{E} + \underline{\underline{\chi}}^{(1)}_{Q} (\underline{\nabla} : \underline{E}) + \underline{\underline{\chi}}^{(2)} \underline{E}\,\underline{E} + \underline{\underline{\chi}}^{(2)}_{Q} \underline{E} (\underline{\nabla} : \underline{E}) \\ & + \underline{\underline{\chi}}^{(3)} \underline{E}\,\underline{E}\,\underline{E} + \underline{\underline{\chi}}^{(3)}_{Q} \underline{E}\,\underline{E} (\underline{\nabla} : \underline{E}) \end{aligned} \qquad (7)$$

or by expressing $\underline{\nabla} : \underline{E}$ in terms of $\underline{\nabla} : \underline{E}$ and $c\underline{\nabla} \wedge \underline{E} = -\dot{\underline{B}}$, the latter obtained from Maxwell equations, expression (7) can be recast in the form

$$\begin{aligned} \underline{P} = {} & \underline{\underline{\chi}}^{(1)} \underline{E} + \frac{1}{c} \underline{\underline{\chi}}^{(1)}_{Q} \dot{\underline{B}} + \underline{\underline{\chi}}^{(2)} \underline{E}\,\underline{E} + \frac{1}{c} \underline{\underline{\chi}}^{(2)}_{Q} \underline{E}\,\dot{\underline{B}} \\ & + \underline{\underline{\chi}}^{(3)} \underline{E}\,\underline{E}\,\underline{E} + \frac{1}{c} \underline{\underline{\chi}}^{(3)}_{Q} \underline{E}\,\underline{E}\,\dot{\underline{B}} \end{aligned} \qquad (8)$$

for an isotropic medium. One can easily see, from the two first terms in (8) together with $\dot{\underline{B}} = -c\underline{\nabla} \wedge \underline{E}$, that one precisely recovers relation (1)

for the linear part of the electric induction. One can also derive the expression of the Poynting vector and that of the energy density.

In the case of the artificial gyrotropic media where a static magnetic field $\underline{H}_0$ is present the polarization can be written

$$\underline{P} = \underline{\underline{\chi}}^{(1)}\underline{E} + \underline{\underline{\chi}}^{(2)}_{M}\underline{E}\,\underline{H}_0 + \underline{\underline{\chi}}^{(2)}\underline{E}\,\underline{E} + \underline{\underline{\chi}}^{(3)}_{M}\underline{E}\,\underline{E}\,\underline{H}_0 + \underline{\underline{\chi}}^{(3)}\underline{E}\,\underline{E}\,\underline{E} + \underline{\underline{\chi}}^{(4)}_{M}\underline{E}\,\underline{E}\,\underline{E}\,\underline{H}_0 \tag{9}$$

Again the two first terms can be recast in a form to recover expression (1) for the linear part of the electric induction. The number of independent components of $\chi^{(n)}_M$ and those of the previously introduced $\chi^{(n)}_Q$ can be derived using the symmetry operations of the medium and have been derived for several symmetry classes. Note that the magnetic fields are not true vectors and care must be paid when applying the space inversion operation.

The quantum mechanical expressions of the susceptibilities $\chi^{(n)}_Q$ and $\chi^{(n)}_M$ can be derived by a perturbative approach starting with the appropriate field-matter interaction terms in the hamiltonian as will be briefly discussed later together with their order of magnitude.

NONLINEAR GYROTROPIC PROPAGATION

The study of the nonlinear propagation in gyrotropic media can be approached by inserting expressions (7) or (8) in the Eq. (3) and applying the slow varying envelope approximation [4] for the field and polarization sources of a given frequency. This approach was first applied [8] for the case of frequency non preserving processes, where the phase mismatch is a key issue, such, as second and third harmonic generation, SHG and THG respectively, and more recently also generalized and extended [9] to the case of optical parametric amplification and oscillation, OPA and OPO respectively, in nonlinear gyrotropic media. In parallel this approach was also extended and applied [10] in the case of frequency preserving processes where the phase mismatch is of lesser importance like intense beam self-action [10–12], degenerate four wave interaction [13], nonlinear gyrotropic resonators and cavities [14–18]. We briefly consider this case here since it concerns the photoinduced gyrotropy; furthermore we restrict

ourselves to the case of artificially gyrotropic media but the case of naturally gyrotropic media can be treated along similar lines.

Starting with (3) and inserting (9) and (1) with $\underline{P}=\underline{P}_L+\underline{P}_{NL}$ we obtain

$$\underline{\nabla}\wedge(\underline{\nabla}\wedge\underline{E})+\ddot{\underline{D}}_L=-4\pi\ddot{\underline{P}}_{NL} \tag{10}$$

where $\underline{D}_L=\underline{E}+4\pi\underline{P}_L$ and $\underline{P}_{NL}$ are the linear and nonlinear parts in (9) respectively and setting

$$\underline{E}=\frac{1}{2}(\hat{e}_+E_+ + \hat{e}E_-)\mathrm{e}^{-i\omega t}+c.c. \tag{11}$$

with $E_\pm = A_\pm \mathrm{e}^{ik_\pm}$ and neglecting phase mismatched terms when applying the slow varying envelope approximation one gets [10, 11] two coupled equations for the two amplitudes

$$\frac{\partial A_+}{\partial z}+\alpha_\pm A_\pm + k'_\pm\frac{\partial A_\pm}{\partial t}+i\frac{k''_\pm}{2}\frac{\partial^2 A_\pm}{\partial t^2}=(\chi\pm\gamma)(|A_\pm|^2+2|A_\mp|^2)A_\pm \tag{12}$$

where $k'_\pm$ and $k''_\pm$ are the inverse group-velocity and chromatic dispersion respectively, $\chi=\omega\chi^{(3)}_{xxxx}/2c$, $\gamma=-3i\omega(\chi^{(4)}_{xyyyz}+\chi^{(4)}_{xxxyz})H_0/8$ and $\alpha_\pm$ are distributed linear loss coefficients. Similar equations have also been derived for the nonlinear propagation of pulses in optical fibers. In the stationary regime without losses these equations reduce to

$$\frac{\partial A_\pm}{\partial z}=(\chi\pm\gamma)(|A_\pm|^2+2|A_\mp|^2)A_\pm \tag{13}$$

These equations have been extended and used to treat the problem of degenerate four wave mixing, and with appropriate boundary conditions to derive the transmission characteristics of nonlinear gyrotropic cavities.

The nonlinear optical propagation in gyrotropic media exhibits some features that are not readily and directly observable in non-gyrotropic media; some of them are specific only to the presence of gyrotropy and can be best assessed in isotropic media. Thus in principle in even isotropic media phase matching in SHG, THG or OPA and other frequency non preserving nonlinear optical processes

can be achieved [8, 9] by compensating the refractive index mismatch at two different frequencies, for instance the fundamental and its harmonics in SHG or THG, with the circular birefringence and using appropriate circularly polarized beams. This was initially proposed [8] for SHG and THG in naturally gyrotropic media where however the circular birefringence is very weak in the optical range to achieve dispersion compensation. More recently this has been extended [9] to the case of artificial gyrotropic media where compensation seems more feasible than in the natural gyrotropic ones with some interesting applications in magneto-optical, optical parametric processes such as MOPG, MOPA and MOPO. For these later cases the Manley–Rowe relations [4] that essentially reflect the flux or electromagnetic momentum (Poynting vector) conversation have been complemented [9] with those of the electromagnetic angular momentum conservation and applied to derive polarization state selection rules for the interacting and generated beams in the optical parametric processes.

In the case of frequency preserving nonlinear optical processes the phase matching problem is less of an issue and a whole class of new types of self-action [10], optical lensing and grating effects can be envisaged. Thus gyrotropic fibers can sustain helicoidal solitary waves [11, 12] of a very robust type; in particular a new scheme for soliton mode locking by nonlinear Faraday rotation has been proposed [11] that in essence exploits the nonreciprocity and the possibility to compensate the linear Faraday rotation with the nonlinear one over a given distance in the fiber, namely when $\theta_{NL} - \theta_R = 0 \bmod (2\pi)$. The case of degenerate four wave interaction has also been treated [13] to some extent in both natural and artificial gyrotropic media in this case. The most striking difference between the two classes of media results from the presence of nonreciprocity in the later case which destroys the time reversal symmetry and the optical phase conjugation properties while these are preserved in the former case where reciprocity is present. This is apparent both in the propagation direction of the signal beam and the ellipticity behavior of its polarization in the two classes of gyrotropic media and also in comparison to non gyrotropic ones. A complication that arises in the nonlinear propagation in gyrotropic media is the decoupling of effects proper to photoinduced circular birefringence (photoinduced gyrotopy) from those to the optical Kerr type photoinduced birefringence; this can be achieved by

proper selection of beam geometry and analysis of the polarization Stokes parameters.

The nonlinear propagation accumulated over multiple reflections in confined gyrotropic media such as gyrotropic resonators [14–18], Fabry Perot cavities, ring cavities and other resonators filled with a gyrotropic medium leads to several strikingly new characteristics regarding transmission, bistable and multistable behavior, instabilities and eventually transition to chaotic behavior. The nonlinear propagation problem in a cavity can actually be treated as a degenerate four wave interaction will all beams collinear and appropriate boundary conditions related to the mirror transmission and reflection. In the case of a gyrotropic ring cavity [15–17] apart from quantitative aspects there is no qualitative difference in the transmission behavior between an artificial and a natural gyrotropic one the main advantage with the artificial one being the possibility to tune the gyrotropic strength and attain the different regimes by changing the magnetic field intensity H_0. Several interesting features have been predicted in this case besides or in conjunction with the bistable behavior. Thus close to threshold values the resonator may covert [16] small changes of the input intensity into large variations of the polarization state and have the cavity act as a polarization state filter; in particular the pitch fork bifurcation can be completely cancelled with small amount of gyrotropy. Also a flip-flop behavior for the polarization state can be obtained by superposing to the input beam an appropriately circular polarized trigger pulse. Finally modulational polarization instability can occur in such a ring cavity.

The case of the nonlinear gyrotropic Fabry Perot cavity [18] consisting of an artificial gyrotopic medium between two parallel mirrors introduces strikingly new features which cannot occur in a Fabry Perot cavity filled with a natural gyrotropic medium. This is because reciprocity is preserved in the later case and in particular the transmission characteristics are not qualitatively much different from those obtained in the ring cavity. In contrast in a nonlinear magnetooptic (artificial gyrotropic) Fabry Perot cavity nonreciprocity is present and manifested in normal reflection; because of the cumulative effect of the multiple reflections and optical nonlinearities then novel features and patterns on the transmission appear. A detailed analysis [18] of the transmission characteristics in terms of the

Stokes parameters revealed that the nonreciprocity in this case leads to specific transmission pattern and, in particular, to a polarization controlled multistability at constant input intensity. It is also effectively switched off/on for certain parameter regimes.

The propagation in nonlinear gyrotropy media still poses some fundamental problems in particular related to the nonlocality and the boundary conditions which will reveal some new regimes in the near future.

NONLINEAR GYROTROPIC POLARIZATION

Along with the problem of the nonlinear optical propagation specific to the gyrotropic media that of the nonlinear gyrotropic polarization source and response in these media is quite intriguing and still not well settled partly due to conceptual difficulties and partly due to still unsettled computational complications as will become shortly evident.

At the outset the nonlinear response of a gyrotropic medium can be obtained [4, 5] by a perturbative treatment of the matter-field interaction as in the case of the nonlinear electric polarization terms in non-gyrotropic media only now besides the electric dipolar interaction term additional ones must be included specific to the gyrotropy. For the case of natural gyrotropy this results [5, 19, 20] in the form

$$\underline{h}' = -\underline{\mu} \cdot \underline{E} - \underline{m} \cdot \underline{B} - \underline{\underline{q}} : \underline{\nabla} \underline{E} \tag{14}$$

for the matter-field interaction where $\underline{\mu}, \underline{m}$ and $\underline{\underline{q}}$ are the electric dipole, magnetic dipole and electric quadrupole operators respectively of the electrons; $\underline{B}$ and $\underline{E}$ are related through the Maxwell equations and we disregard here the distinction [5] between local (microscopic) and macroscopic fields which may affect at the final stage the magnitude of the coefficients. Within the physical context we restrict ourselves here and exemplified by the form of the macroscopic polarization (7), in the perturbative treatment the electric dipole term $\underline{\mu} \cdot \underline{E}$ is treated up to the requested order in the different terms in (7) while the other two terms in (14) are kept only to first order. Concommitantly one must also set up the induced magnetic and quadrupolar polarization densities as

well, $\underline{M}$ and $\underline{\underline{Q}}$ respectively, but the relevant coefficients are related by certain permutation symmetries with those of the electric polarization density (7) and we need not discuss them explicitly here. It is then quite evident to see how nonlocality creeps [5] into natural gyrotropy namely through the presence of spatial derivatives of the electric oscillating field $\underline{E}$ and also the presence of the oscillating magnetic field related to the electric one through Maxwell's equation $\dot{\underline{B}} = -c\underline{\nabla} \wedge \underline{E}$. In particular if the plane wave Fourier representation of the fields is used these derivatives introduce a $\underline{k}$-dependence or equivalently wavelength dependence in the susceptibilities, the so called spatial dispersion [21], another manifestation of nonlocality and closely connected with natural gyrotropy or natural optical activity. The formal quantum mechanical expressions of the gyrotropic nonlinear susceptibilities have been derived [20] for different processes but their computation has not been attempted neither their behavior close to resonances has been studied to any extent.

For the case of the artificial gyrotropy the relevant interaction hamiltonian [4, 19] is

$$\underline{h}' = -\underline{\mu} \cdot \underline{E} - \underline{m} \cdot \underline{H}_0 \tag{15}$$

where now the static magnetic field $\underline{H}_0$ bears no relation to the oscillating electric or magnetic field of the e.m. field. Referring to the macroscopic polarization (9) the different terms there can be obtained by a perturbative treatment of (15) keeping terms up to the requested order in the electric dipole interaction $\underline{\mu} \cdot \underline{E}$ but only linear in $\underline{m} \cdot \underline{H}_0$. The time reversal symmetry here breaks down and nonreciprocity is introduced; furthermore the susceptibility coefficients satisfy [1, 22]. Onsager type relations. The relevant susceptibilities have been derived [4, 23] for several magnetooptical processes but here again their computation and resonance behavior have not been studied to any extent.

At the qualitative level the magnetooptical effects are relatively well understood but their quantitative prediction is lagging or missing. Most of the effects related to the different magnetooptical polarization terms and their inverse have been observed and some cases are exploited in applications or are in wait. Thus the Faraday effect is related to the polarization source $\chi_M^{(2)} EH_0$ which can be related to the gyration vector $\underline{g}$ in the linear part of (1); $\chi_M^{(2)} EE^*$ is related to the

inverse Faraday effect or photoinduced magnetization, in a certain way the analogue of the optical rectification effect. The photoinduced Faraday effect [7] is related to $\chi_M^{(4)} EE^* E H_0$ and clearly one can have the inverse effect in $\chi_M^{(4)} EE^* EE^*$ which is an additional contribution to the photoinduced magnetization.

These later effects are however relatively weak unless one is close to a resonance and also makes provisions to enhance the magnetooptic coupling. The first situation can be encountered in the narrow atomic resonances [24, 25] of atomic vapors where for the first time such effects were observed or in the narrow and intense excitonic transitions [26] in quantum confined nanostructures of semimagnetic semiconductors where in addition the magnetooptic coupling can be enhanced. In the later case, following previous studies in bulk compounds [27] indeed giant photoinduced Faraday rotations have been evidenced [26] close to such excitonic transitions as well as photoinduced magnetization with circularly polarized light in the absence of an external magnetic field. Besides the impact of the resonance the enhancement here mainly results from the specific spin-exchange interaction $J\underline{s} \cdot \underline{S}$ between the delocalized *sp*-type band electrons and the magnetic impurity localized *d*-type electrons in these materials [27]. This effectively results in a two-orders of magnitude temperature dependent enhancement of the Landé factor g from its bare electron value $g_0 = 2$. The underlying mechanism [26] in this giant photoinduced Faraday rotation is the polarization state selective saturation of the Zeeman components of the excitonic transition in the presence of a magnetic field; the photoinduced Faraday rotation angles can be as large as those of the linear Faraday rotation and in fact cancel the later namely they can reach values of the order of 20 degrees over a 1 μm thick multiquantum well nanostructure in moderate magnetic fields ($\sim 2T$) and moderate light intensities. In the absence of the magnetic field circularly polarized light of comparable intensity can induce magnetization of up to $0.5\,T$ in the same structure. Several additional processes were identified to contribute to these photoinduced gyrotropy and magnetization. One can envisage several applications of these effects in particular non-reciprocal light valves and unidirectional shielding of optical information.

The corresponding effects in naturally gyrotropic media such as optically active media or chiral molecular systems are much harder to

evidence because of the inherent nonlocality in the matter-field interaction which complicates the description of the processes and also because of the smallness of the relevant coefficients in the optical range even close to resonances. For instance photoinduced optical activity [28] in chiral molecules such as helicenes hardly exceeds a few minutes of a degree in a high concentration sample of 1 cm length and intense light beams. The relevant physical parameter in the natural gyrotropy is the Rosenfeld rotational strength [29, 19] $R_{ng} = \mathrm{Im}(\underline{\mu}_{gn} \cdot \underline{m}_{ng})$ of an electric and magnetic dipole allowed transition $(g \rightarrow n)$; this is in essence a mixed vector product, since $\underline{m}$ is a pseudovector, and represents an oriented quantum volume element imposing quite specific restrictions on the electronic level configuration since these strenghts must satisfy the sum rule $\Sigma_n \mathrm{Im}(\underline{\mu}_{gn} \cdot \underline{m}_{ng}) = 0$. Thus this implies that the relevant transitions occur in pairs partially cancelling each other in this sum with the rotational strengths reversing sign when going from one enantiomer to the other while their magnitudes are preserved; this is a consequence of the stereochemical structure of these molecules. To some extend the magnitude and sign of these rotational strengths can be determined by measuring the circular dichroism of the molecules and can give [6, 19] important information regarding the stereochemical structure of these molecules; polarization sensitive nonlinear optical techniques can provide [30–32] additional information in particular regarding rotational strengths between excited states and vibrational dynamics which cannot be accessed with the conventional circular dichroism measurements.

There is growing interest in the study of nonlinear gyrotropic effects on surfaces and interfaces. Both natural gyrotropic surfaces and interfaces such as chiral organic ones [33, 34] and artificial gyrotropic ones such as thin magnetic films [35, 36] with modulated magnetization present are being studied with polarization state and surface symmetry sensitive nonlinear optical techniques. The most promising technique is that of the reflection of the second harmonic and sum frequency which is very sensitive to the breakdown of inversion and mirror reflection symmetries but higher even order processes can also be envisaged. There are several interesting fundamental aspects related to such studies with several competing mechanisms contributing there and satisfying certain selection rules. The extension of such studies and the understanding of these mechanisms in the case of chiral surfaces

and interfaces can lead to the development of powerful techniques to study and control such important aspects as growth of chiral supramolecular or crystalline complexes that play an important role in biology, biochemistry, drug development, asymmetric catalysis and molecular recognition. In the case of magnetic interfaces and surfaces such studies can lead to better understanding of the surface magnetism, in particular that of the last layer of magnetic materials, and the magnetooptic recording techniques.

EVALUATION OF THE GYROTROPIC SUSCEPTIBILITIES

The quantummechanical expressions of the nonlinear gyrotropic susceptibilities have been derived [4, 37, 20, 17] for practically all envisageable nonlinear gyrotropic processes with the complete frequency and transition strength dependencies well exhibited. There have been no attempts to estimate these coefficients either in a transparency region or close to a resonance and this situation is actually true with few important exceptions even for the case of the relatively much simpler linear gyrotropic coefficients namely the linear rotatory power and the Verdet constant for the natural and artificial gyrotropy respectively. The few important exceptions concern the estimation of the rotatory power and circular dichroism coefficients of certain optically active organic molecules and chiral polymeric chains either conjugated or saturated. Here we do not count the extensive studies of circular dichroism of biological organic molecules used for the determination of certain stereochemical structural aspects not directly transferable to the estimation of the relevant gyrotropic coefficients.

The difficulty in estimating the gyrotropic coefficients stems from their complex structure involving both electric dipole and magnetic dipole transition elements, in contrast to the electric dipolar susceptibilities that involve only electric dipole transition elements, and from other very subtle three-dimensional aspects namely the oriented "quantum volume" transition elements such as $\mathrm{Im}(\underline{\mu}_{gn} \cdot \underline{m}_{ng})$ involved there. These together with the sums-over-states that appear in these coefficients require very complex calculations with very accurate

wave functions both in regard to amplitude and phase. In fact these coefficients are in general complex numbers even away from resonances in contrast to the electric dipolar susceptibilities which are real and essentially only sensitive to the amplitude of the wave function (or equivalently, the electronic charge density distribution). In particular the calculation of the matrix elements of $\underline{m}$ or $\underline{L}$ involves derivatives of the wave functions that appreciably amplify or suppress certain features and are therefore very sensitive to errors or inaccuracies there. The situation is somewhat better in the case of magnetooptic coefficients but still the estimations even here have not been developed to any extent.

Keeping well in sight the present lack and future need of consistent and physically acceptable calculations of the nonlinear gyrotropic coefficients we propose here an order of magnitude estimation based on dimensional scaling arguments and comparison of the electrical dipolar susceptibilities and the gyrotropic ones of the same order in the electric field amplitude. By referring to their quantum mechanical expressions we expect that

$$\frac{\Delta\chi^{(3)}}{\chi^{(3)}} \simeq \frac{\Delta\chi^{(1)}}{\chi^{(1)}} = \sigma$$

where $\Delta\chi^{(1)}$ and $\Delta\chi^{(3)}$ are the coefficients that describe the gyrotropic polarization terms linear and cubic in the electric field amplitudes respectively: in the case of the natural gyrotropy $\Delta\chi^{(1)} = k\chi_Q^{(1)}$ and $\Delta\chi^{(3)} = k\chi_Q^{(3)}$ and in the case of the artificial gyrotropy $\Delta\chi^{(1)} = \chi_M^{(2)} H_0$ and $\Delta\chi^{(3)} = \chi_M^{(4)} H_0$ respectively. The ratio $\Delta\chi^{(1)}/\chi^{(1)}$ can be easily determined experimentally by measuring the polarization rotation angle of a linearly polarized optical beam and expressing it in terms of the optical rotatory power or Verdet constant. One can extent this approach also close to a resonance. Thus for the photoinduced Faraday rotation in a glass material with $V = 10\,[\text{rad}\ T^{-1}m^{-1}]$ and $n_0 = 1.5$ at $\lambda = 1.5\,\mu\text{m}$ we find $\sigma = \Delta\chi^{(3)}/\chi^{(3)} = 10^{-5}$ for $H_0 = 1\ T$ on the other hand for the case of photoinduced optical activity we expect $\chi\chi^{(3)}/\chi^{(3)} \approx 10^{-4}$ at $\lambda \approx 1.5\,\mu\text{m}$, for a medium with $n_0 = 1.5$. This order of magnitude estimation seems to be corroborated by some recent attempts for more accurate estimation of these coefficients.

GENERAL REMARKS

The optical gyrotropy and its modification by nonlinear optical interactions is connected with some very fundamental problems in matter-field interaction and propagation with far reaching applications that only recently started emerging. The relevant effects and in particular the photoinduced gyrotropy can be appreciable and easily discriminated against the background of other nonlinear processes. Although still far-fetched some of these aspects and concepts may also have [38] an impact in recent considerations on quantum mechanical measurements. There are striking differences on the nonlinear gyrotropic behavior of artificial and natural gyrotropic materials. In the former class the inherent nonreciprocity of the magnetooptic coupling can lead to conceptually novel applications regarding unidirectional control or shielding of optical signal transfer, or storage and transfer of coherence, quantum optical or spin coherence. The latter is reciprocal but very sensitive to the nonlocal character of the feld-matter interaction and the nonlinear optical gyrotropic effects there can be exploited to develop sensitive diagnostic techniques of chiral molecular systems in particular dynamical processes that can take place there and their interactions.

We expect that the growing study of nonlinear gyrotropic interactions will lead to some new and interesting fundamental and technological considerations.

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