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MEASURABLE MAGNETO-OPTICAL CROSS EFFECTS

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Laser light induces measurable variations in Faraday and Cotton-Mouton effects in isotropic media, particulary large in macromolecular and colloidal substances.

Present laser techniques of producing intense optical fields ($E \sim 10^6$ esu in a focused beam) as well as pulse methods of producing strong magnetic fields ($H \sim 10^6$ Oe) permit one to induce in isotropic bodies various magneto-optical higher-order effects which, as will be shown here, are accessible to experimental detection.

In any medium, the electric field $E \exp(i\omega t) +$ c.c. of a measuring light beam oscillating at frequency ω will induce electric polarization, which in the dipolar approximation is

$$P_\sigma(\omega) = \chi_{\sigma\tau}(\omega)\, E_\tau(\omega) \tag{1}$$

Moreover, when the medium is immersed in a strong dc magnetic field, H, the electric susceptibility tensor $\chi_{\sigma\tau}$ becomes a function of H. In the linear and quadratic approximations, respectively,

$$\chi^{(1)}_{\sigma\tau}(\omega) = \chi^{em}_{\sigma\tau\nu}(\omega) H_\nu\,, \tag{2}$$

$$\chi^{(2)}_{\sigma\tau}(\omega) = \chi^{em}_{\sigma\tau\nu\rho}(\omega) H_\nu H_\rho\,. \tag{3}$$

If the medium is isotropic, Eq. (2) leads to magneto-optical rotation[1] (the light beam propagating along the z axis, see Fig. 1):

$$n^{(1)}_{xy} - n^{(1)}_{yx} = F_0(\omega) H_z\,, \tag{4}$$

$F_0(\omega) = \pi\chi^{em}_{\alpha\beta\gamma}(\omega)\epsilon_{\alpha\beta\gamma}/3n$ being Faraday's constant, whereas Eq. (3) yields the well-known magneto-optical birefringence[2]:

$$n^{(2)}_{xx} - n^{(2)}_{yy} = nC(\omega)\,(H_x^2 - H_y^2), \tag{5}$$

where $C(\omega) = \pi\chi^{em}_{\alpha\beta\gamma\delta}(\omega)\,(3\delta_{\alpha\gamma}\delta_{\beta\delta} + 3\delta_{\alpha\delta}\delta_{\beta\gamma} - 2\delta_{\alpha\beta}\delta_{\gamma\delta})/15n^2$ is the Cotton-Mouton constant; $\delta_{\alpha\beta}$ is the unit Kronecker tensor and $\epsilon_{\alpha\beta\gamma}$ the Levi-Cività pseudotensor and n-refractive index.

We shall now consider the case when the medium is, moreover, acted on by a laser field $E_L \exp(i\omega_L t) +$ c.c oscilating at frequency ω_L of intensity sufficient for causing additional, nonlinear polarization. We now get, in the second approximation,

$$\chi^{(2)}_{\sigma\tau}(\omega) = \chi^{ee}_{\sigma\tau\nu\rho}(\omega, \omega_L) E_\nu(\omega_L) E_\rho(-\omega_L), \tag{6}$$

—an expression leading to the optical Kerr effect

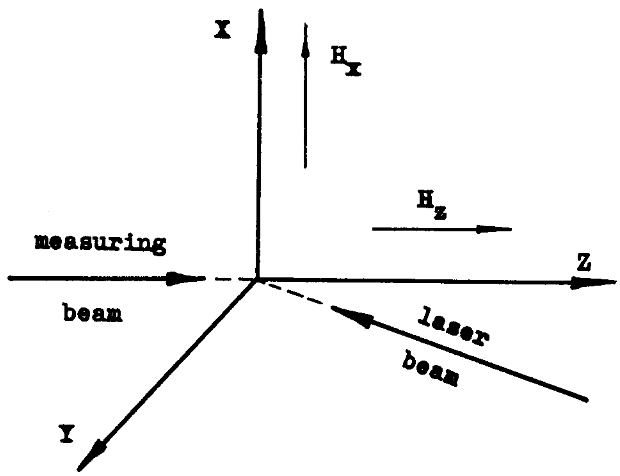

Fig. 1. Proposed experiments, diagramatically. The measuring beam, conveying the electric field E, propagates in the direction of the Z axis. The dc magnetic field H acts along the Z axis (Faraday's effect) or in the XY plane (Cotton-Mouton effect). The intense laser beam with electric field E_L can propagate in any direction, but maximal values of the magneto-optical cross effects are obtained when it propagates along the Z axis.

already studied experimentally in liquids[3,4]:

$$n_{xx}^{(2)} - n_{yy}^{(2)} = nK(\omega, \omega_L)\{E_x(\omega_L)E_x(-\omega_L) - E_y(\omega_L)E_y(-\omega_L)\} \quad (7)$$

with the constant $K(\omega, \omega_L) = \pi\chi^{ee}_{\alpha\beta\gamma\delta}(\omega, \omega_L)\ (3\delta_{\alpha\beta}\delta_{\beta\delta} + 3\delta_{\alpha\delta}\delta_{\beta\gamma} - 2\delta_{\alpha\beta}\delta_{\gamma\delta})/15n^2$.

In addition to these independent quadratic effects, on proceeding to higher approximations one obtains various magneto-optical cross effects. Here, we shall discuss only the more important ones.

In the third approximation, we have the expression:

$$\chi^{(3)}_{\sigma\tau}(\omega) = \chi^{em}_{\sigma\tau\nu\rho\lambda}(\omega, \omega_L)E_\nu(\omega_L)E_\rho(-\omega_L)H_\lambda\,, \quad (8)$$

which yields

$$n_{xy}^{(3)} - n_{yx}^{(3)} = F_1(\omega, \omega_L)\{E_x(\omega_L)E_x(-\omega_L) + E_y(\omega_L)E_y(-\omega_L)\}H_z \quad (9)$$

for the laser-induced variation in magneto-optical rotation, with Faraday constant given as

$$F_1(\omega, \omega_L) = \pi\chi^{em}_{\alpha\beta\gamma\delta\epsilon}(\omega, \omega_L)\{2\epsilon_{\alpha\beta\epsilon}\delta_{\gamma\delta} + \epsilon_{\alpha\gamma\epsilon}\delta_{\beta\delta} + \epsilon_{\alpha\delta\epsilon}\delta_{\beta\gamma} - \delta_{\alpha\gamma}\epsilon_{\beta\delta\epsilon} - \delta_{\alpha\delta}\epsilon_{\beta\gamma\epsilon})/15n.$$

By analogy to the case of light-intensity-dependent refractive index,[4,5] one can study the nonlinear variations in Faraday constant caused by a single, very intense laser beam.[6]

Going over from the preceding phenomenological considerations to the molecular-statistical picture,[2] one can show that for a medium of anisotropic molecules the ratio F_1/F_0 is of the order $\alpha/kT = 10^{-10}$ (with α the linear optical polarizability) permitting one to detect nonlinearities of the Faraday effect at laser field strengths of the order of 10^4–10^5 esu. In addition to this temperature effect resulting from molecular reorientation in the electric laser field, the constant F_1, moreover, contains a contribution from nonlinear electronic polarizability, which is nonzero even for molecules of high symmetry (thus, of point groups T_d, T, 0_h or 0). Albeit, this temperature-independent effect is small, yielding F_1/F_0 of the order of 10^{-14} or, at the most, 10^{-12}.

Significantly, calculations show that the reorientation effect can be particularly large in macromolecular or colloidal substances. For example, the optical Kerr constant of gold-water colloid[7] is by three orders of magnitude larger than in pure water.[3] Indeed, the polarizability of gold particles of diameter 300 Å amounts to 10^{-18} cm^3 for λ = 6500 Å leading to F_1/F_0 of the order 10^{-4}, whence in colloidal systems the variations in Faraday constant are seen to be measurable already at laser field strengths of 100 esu.

In the fourth approximation, there appears a cross effect dependent simultaneously on the square of the laser field and the square of the dc magnetic field, namely:

$$\chi^{(4)}_{\sigma\tau}(\omega) = \chi^{em}_{\sigma\tau\nu\rho\lambda\mu}(\omega, \omega_L)E_\nu(\omega_L)E_\rho(-\omega_L)H_\lambda H_\mu\,. \quad (10)$$

For a probe beam propagating along H, one obtains:

$$n_{xx}^{(4)} - n_{yy}^{(4)} = nC_1(\omega, \omega_L)\{E_x(\omega_L)E_x(-\omega_L) - E_y(\omega_L)E_y(-\omega_L)\}H_z^2\,, \quad (11)$$

$$n_{xy}^{(4)} - n_{yx}^{(4)} = F_2(\omega, \omega_L)\{E_x(\omega_L)E_y(-\omega_L) - E_y(\omega_L)E_x(-\omega_L)\}H_z^2\,, \quad (12)$$

the constants C_1 and F_2 being given by appropriate combinations of components $\chi^{em}_{\alpha\beta\gamma\delta\epsilon\eta}$. Yet other effects result for a probe beam propagating perpendicularly to H; thus, we get birefringence

$$n_{xx}^{(4)} - n_{yy}^{(4)} = nC_2(\omega, \omega_L)E_x(\omega_L)E_x(-\omega_L)H_x^2 \quad (13)$$

if $E(\omega_L)$ oscillates parallel to H. The constant C_2, which is a new contribution to the usual Cotton-Mouton effect (Eq. 5), is of the order of $(\alpha/kT)E_x^2$ for anisotropic molecules and can easily be disclosed in colloidal systems by laser technique. Also, one can regard Eq. (13) as a contribution to the optical birefringence of Eq. (7) due to the square of H. For diamagnetic molecules, it is $\sim 10^{-14} H_x^2$ and begins to play a part at $H \sim 10^6$ Oe. Obviously, in colloidal systems this effect is by 4–6 orders of magnitude larger, and is easily observable already at magnetic fields of 10^4–10^5 Oe, such as are applied when measuring the Cotton-Mouton effect in liquids.[8]

Similarly, one can discuss the variations induced in the magnetic susceptibility tensor by strong fields $E(\omega_L)$ and H. In particular, a quadratic approximation will yield expressions like Eq. (7) accounting for the magnetic anisotropy induced in isotropic media by intense laser light.[9] Likewise, one can calculate magnetic contributions to the nonlinear Faraday effect which are, however, insignificant.

The magneto-optical effects discussed above in a phenomenological approach can be uniformly treated quantum mechanically[10,11] or by semimacroscopic or molecular-statistical[2,9] methods, which provide insight into their microscopic mechanisms, with the aim of deriving novel information regarding the strongly nonlinear electromagnetic properties of molecules and the structure of media. It is hoped that present work at this Department on intense laser light propagation in media acted on by dc magnetic fields will help to further clarify nonlinear optical processes,[5,12] including self-trapping.[13]

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