NATURAL AND MAGNETO-OPTICAL ROTATION IN THE PRESENCE OF AN INTENSE LIGHT BEAM

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By classical theory it is shown that light intensity-dependent changes in natural and magneto-optical rotation should be accessible to observation by appropriately applied available laser techniques.

Extending the usual optical rotation theory of dense isotropic media [1] to the case of an intense light beam of intensity I, we obtain the following change in optical permittivity tensor (omitting the diagonal part accounting for the optically induced birefringence discussed previously [2]):

$$\Delta n^2_{\sigma\tau} = (A_0 + A_1 I + A_2 I^2 + \ldots)\, \epsilon_{\sigma\tau\nu} k_\nu ; \qquad (1)$$

k_ν is the light wave propagation vector component and $\epsilon_{\sigma\tau\nu}$ the Levi-Civitã extensor.

The constant A_0 accounts for natural optical rotation in the absence of the intense beam

$$A_0 = \frac{2\pi i}{3V}\left(\frac{n^2+2}{3}\right)\langle B_{\alpha\beta\gamma}\,\epsilon_{\alpha\beta\gamma}\rangle ; \qquad (2)$$

the extensor $B_{\alpha\beta\gamma}$ defines the optical polarizability tensor of the medium of volume V due to the gradient of the optical electric field; the symbol $\langle\quad\rangle$ denotes appropriate statistical averaging.

The constants $A_1, A_2, \ldots$ in (1) describe departure from usual optical rotation (2) due to strong light. The explicite form of A_1 is

$$1 = \frac{\pi i}{30V}\left(\frac{n^2+2}{3}\right)\langle\{D_{\alpha\beta\gamma\delta\epsilon} + \beta B_{\alpha\beta\gamma}(A_{\delta\epsilon} - \langle A_{\delta\epsilon}\rangle)\}\,\epsilon_{\alpha\beta\gamma\delta\epsilon}\rangle , \qquad (3)$$

where $\beta = 1/kT$ and the extensor $\epsilon_{\alpha\beta\gamma\delta\epsilon}$ consists of $\delta_{\alpha\beta}$ and $\epsilon_{\gamma\delta\epsilon}$.

The tensors $A_{\alpha\beta}$, $B_{\alpha\beta\gamma}$ and $D_{\alpha\beta\gamma\delta\epsilon}$ which characterize the macroscopic optical properties of the medium, can obviously be expressed in terms of appropriate polarizability tensors of individual molecules. As we see, A_1 consists of two terms; the first, temperature-independent results from the nonlinear change in extensor $A_{\alpha\beta\gamma}$ due to the square of the optical electric field, whereas the second temperature-dependent term arises from the statistical molecular orientation effect, with $A_{\alpha\beta}$ denoting the linear polarizability tensor.

Expression (3) holds for molecules of orbitrary symmetry. For spherically symmetric molecules the constant (2) and the temperature-dependent term in (3) vanish, but not the temperature-independent term, which exist owing to the activity induced in a spherical molecule by the strong optical field. This is the case of optically induced optical rotation. In the absence of molecular interaction in the medium the temperature-dependent term in (3), like the constant (2), exists only for optically active molecules.

In the case of incidence along the Z axis, we obtain by (1)

$$n_R^2 - n_L^2 = 2(A_0 + A_1 I + A_2 I^2 + \dots) k_z \qquad (4)$$

for the difference in refractive indices for right and left circularly polarized light.

Since for anisotropic inactive molecules the temperature-dependent term in A_1 is predominant, the ratio A_1/A_0 is of the order of 10^{-10}; hence by (4) the change in $n_R - n_L$ due to intense light is of the order of $10^{-10} I$ and is accessible to measurement at $I \simeq 10^5$ esu. It is noteworthy that measurements of a similar effect in some organic liquids have been performed by laser techniques [3].

Similarly, applying the semi-macroscopic theory of magneto-optical phenomena [4] we obtain for the case when the isotropic medium is acted on simultaneously by a weak magnetic field H and a strong optical electric field:

$$\Delta n_{\sigma\tau}^2 = (F_0 + F_1 I + F_2 I^2 + \dots) \epsilon_{\sigma\tau\nu} H_\nu . \qquad (5)$$

Here, F_0 is the Faraday's constant in the absence of intense light, whereas the constants F_1, $F_2, \dots$ account for the change in magneto-optical rotation due to strong light. The explicite form of F_1 is analogous to (3) and, on certain assumptions, can be simplified to a form resembling the constant D discussed recently by Graham and Raab [5] in their theory of the strong magnetic field Faraday effect in diamagnetic gases.

Numerical evaluations of A_1/A_0 and F_1/F_0 show that in some optically inactive substances present laser techniques are adequate for the detection of light intensity-dependent changes, both in natural and magneto-optical rotation. Investigation of these new nonlinear optical processes will provide information on the structure of individual molecules as well as on the forces with which they interact in dense media.

References

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