DOUBLING AND MIXING OF LASER LIGHT FREQUENCIES IN CROSSED ELECTRIC AND MAGNETIC FIELDS

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Processes of second harmonic generation and laser wave mixing in isotropic bodies in crossed electric and magnetic fields E^0 and H^0 are analyzed quantitatively. Suitable experimental set-ups are devised for the study of these hitherto undiscussed magneto-electro optical processes yielding the 6 independent components of the new fifth-rank pseudo-tensor $\chi_{ijklm}(-\omega_3, \omega_1, \omega_2, 0, 0)$ defining the changes induced in the susceptibility tensor $\chi_{ijk}(-\omega_3, \omega_1, \omega_2)$ by coupled fields E^0 and H^0, and providing valuable data on the structure of matter and microsystems.

The second-order polarisation component $P_i(\omega_3)$ induced in a medium at frequency $\omega_3 = \omega_1 + \omega_2$ by two mutually interacting fields $E_j^{\omega_1}$ and $E_k^{\omega_2}$ oscillating at frequencies ω_1 and ω_2 is given by [1]:

$$P_i(\omega_3) = \chi_{ijk}^{\omega_3} E_j^{\omega_1} E_k^{\omega_2} . \quad (1)$$

The third-rank tensor elements $\chi_{ijk}^{\omega_3} = \chi_{ijk}(-\omega_3, \omega_1, \omega_2)$ of the second-order nonlinear susceptibility are non-zero in the absence of a centre of symmetry (summation is over recurring indices j,k labelling the x,y,z laboratory axes).

In naturally isotropic bodies, the centre of symmetry is removed by applying a dc electric field E^0, and the electric-dipole second harmonic of light is intensely generated even in molecular substances [2]; moreover, frequency mixing can take place [3]. If the dc field is strictly uniform and if the dependence of $\chi_{ijk}^{\omega_3}$ on E^0 is linear [3], SHG takes place only if the wave is made to propagate perpendicularly to the lines of force of the field E^0 [2]. The situation changes when the electric field is inhomogeneous, or if the body is naturally gyrotropic (optically active) or when gyrotropy is induced in it by a magnetic field as is the case in Faraday's (linear and nonlinear) effect [4].

The present analysis will bear on the case of magnetic gyrotropy, when an isotropic body is acted on with two crossed fields concomitantly, namely an electric field E^0 and a magnetic field H^0 (fig. 1). In this experimental set-up, the polarisation [1] and thus the tensor $\chi_{ijk}^{\omega_3}$ become functions of the field strengths E^0 and H^0:

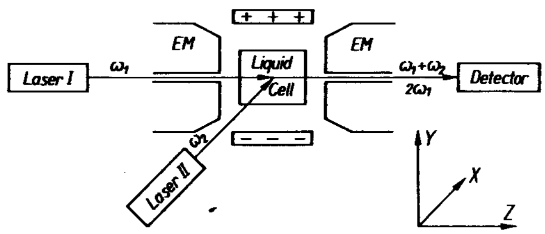

Fig. 1. Scheme of experimental setup for the study of frequency doubling and mixing of two laser waves E^{ω_1} and E^{ω_2} traversing a liquid cell placed between crossed electric and magnetic fields E_y^0 and H_z^0.

$$\chi_{ijk}^{\omega_3}(E^0, H^0) = \chi_{ijklm}^{\omega_3} E_l^0 H_m^0 . \quad (2)$$

The pseudo-tensor $\chi_{ijklm}^{\omega_3} = \chi_{ijklm}(-\omega_3, \omega_1, \omega_2, 0, 0)$ of rank 5 defines the nonlinear change of the tensor $\chi_{ijk}^{\omega_3}$ caused by the crossed fields E^0 and H^0 jointly.

In the case of the isotropic body, the pseudotensor $\chi_{ijklm}^{\omega_3}$ reduces its 243 elements to 60 non-zero ones, 6 of which are in general mutually independent. Assuming a situation in which the dc electric field and the electric fields of the waves are perpendicular to the magnetic field, we have by averaging the fifth-rank pseudotensor over all possible directions with equal probability [4]:

$$\chi^{\omega_3}_{ijklm} = \chi^{\omega_3}_1 \delta_{ij}\epsilon_{klm} + \chi^{\omega_3}_2 \delta_{ik}\epsilon_{jlm} + \chi^{\omega_3}_3 \delta_{jk}\epsilon_{ilm} + \chi^{\omega_3}_4 \delta_{il}\epsilon_{jkm} + \chi^{\omega_3}_5 \delta_{jl}\epsilon_{ikm} + \chi^{\omega_3}_6 \delta_{kl}\epsilon_{ijm} , \quad (3)$$

with the constants $\chi^{\omega_3}_1, \dots, \chi^{\omega_3}_6$ expressed in terms of the relevant tensor elements $\chi^{\omega_3}_{ijklm}$ whereas δ_{ij} and ϵ_{ijk} are the symmetric Kronecker unit tensor and the antisymmetric Levi-Cività tensor.

Once we can tell from eq. (3) which of the tensor elements $\chi^{\omega_3}_{ijklm}$ are non-zero, we can find the corresponding components of second-order polarisation (1). Let us assume crossed fields as shown in fig. 1, where the dc electric field E^0 acts along y and the magnetic field H^0 along z. Eqs. (1)-(3) now yield:

$$P_x(\omega_3, E^0_y, H^0_z) = \left\{ \chi^{\omega_3}_{xxxyz} E^{\omega_1}_x E^{\omega_2}_x + \chi^{\omega_3}_{xyyyz} E^{\omega_1}_y E^{\omega_2}_y + \chi^{\omega_3}_{xzzyz} E^{\omega_1}_z E^{\omega_2}_z \right\} E^0_y H^0_z ,$$

$$P_y(\omega_3, E^0_y, H^0_z) = \left\{ \chi^{\omega_3}_{yxyyz} E^{\omega_1}_x E^{\omega_2}_y + \chi^{\omega_3}_{yyxyz} E^{\omega_1}_y E^{\omega_2}_x \right\} E^0_y H^0_z , \quad (4)$$

$$P_z(\omega_3, E^0_y, H^0_z) = \left\{ \chi^{\omega_3}_{zxzyz} E^{\omega_1}_x E^{\omega_2}_z + \chi^{\omega_3}_{zzxyz} E^{\omega_1}_z E^{\omega_2}_x \right\} E^0_y H^0_z ,$$

and the following symmetry relations hold:

$$\chi^{\omega_3}_{xxxyz} = \chi^{\omega_3}_{xyyyz} + \chi^{\omega_3}_{yxyyz} + \chi^{\omega_3}_{yyxyz} = \chi^{\omega_3}_{xzzyz} + \chi^{\omega_3}_{zxzyz} + \chi^{\omega_3}_{zzxyz} . \quad (5)$$

A further reduction in number of independent elements (3) and (5) is achieved on neglecting dispersion and absorption and on resorting to other symmetry relations.

The expressions (4) suggest certain experimental setups permitting numerical determinations of the various tensor elements [5].

i) Let the two waves propagate along the z axis parallel to one another (in the same or opposite directions) i.e. along the magnetic field H^0, as in the nonlinear Faraday effect [4]; by measuring $P_x(\omega_3)$ and $P_y(\omega_3)$, one will determine the tensor elements $\chi^{\omega_3}_{xxxyz}, \chi^{\omega_3}_{xyyyz}, \chi^{\omega_3}_{yxyyz}, \chi^{\omega_3}_{yyxyz}$. In particular SHG can take place and, with the incident light beam linearly polarized, the element $\chi^{2\omega}_{xyyyz}$ can be determined at oscillations along E^0_y (from the component $P_x(2\omega)$ given by (4) for $\omega_1 = \omega_2 = \omega$); if the oscillations are perpendicular to E^0_y and parallel to x, one gets the element $\chi^{2\omega}_{xxxyz}$. Using circularly polarized light one can moreover measure the component $P_y(2\omega)$, which enables us to determine the elements $\chi^{2\omega}_{yxyyz}$ and $\chi^{2\omega}_{yyxyz}$.

ii) Another situation would be with both waves propagating along the lines of force of the field E^0_y (e.g. by making them traverse condenser plates transparent for the wavelengths used) perpendicularly to the magnetic field H^0_z. Measurement of the components $P_x(\omega_3)$ and $P_z(\omega_3)$ will now permit the determination (similarly as above) of the elements $\chi^{\omega_3}_{xxxyz}, \chi^{\omega_3}_{xzzyz}, \chi^{\omega_3}_{zxzyz}, \chi^{\omega_3}_{zzxyz}$.

iii) The two waves are incident along x i.e. perpendicularly to the plane yz in which the fields E^0_y and H^0_z act. If their oscillations are parallel to the y axis or z axis only, all polarisation components (4) will vanish. In this setup, nonlinear processes can occur only if the oscillation planes of the waves subtend an angle with the directions of the lines of force of E^0_y and H^0_z.

iv) A frequency-mixing situation can be imagined where the two light beams intersect one another with, say, E^{ω_1} propagating along the lines of force of the magnetic field and E^{ω_2} at some angle to or parallel to E^0 (fig. 1). In these conditions, the elements $\chi^{\omega_3}_{xxxyz}, \chi^{\omega_3}_{zxzyz}, \chi^{\omega_3}_{yyxyz}, \chi^{\omega_3}_{zxzyz}$ can be determined, according to the state of polarisation of the waves.

Obviously, the expressions (4) provide the basis for a discussion of yet other geometrical configurations between the propagation directions k_1 and k_2 of the waves, their polarisations, and the directions of E^0 and H^0, permitting the observation of a sum-frequency polarisation wave with propagation vector $k_3 = k_1 + k_2$. Our considerations are easily extended to cases of wave mixing in materials of other symmetries, thus cubic crystals and other weakly nonlinear materials [5], as well as liquid crystals [6].

The above discussion shows that we have at our disposal the experimental possibility of determining all 6 independent elements of the pseudo-tensor $\chi^{\omega_3}_{ijklm}$, which describes not only the frequency doubling and mixing processes dealt with here but moreover the nonlinear changes in Verdet constant induced in Faraday's effect by an electric or optical field [4,7]. The phenomenological treatment used here can be replaced by a quantum-mechanical or statistical-molecular approach permitting insight into the microscopic mechanisms of these nonlinear processes which, obviously, hinge primarily on processes of nonlinear electron distortion of microsystems (atoms, molecules, ions, macromolecules, colloid particles) described by the fifth-rank pseudo-tensor of nonlinear molecular polarisability discussed recently by Boyle [8] for all point groups. In general, processes of reorientation and rearrangement of the microsystems will not be negligible [9], particularly at low temperatures and in strongly condensed states. The new experimental situations which we propose here for the study of induced wave-mixing processes should prove higly feasible in macromolecular and colloidal solutions [4,7,9] and in liquid crystals [6]. We surely have here a source of new information regarding the nonlinear optical properties of microsystems and their nonlinear variations induced by the joint, cross action of electric and magnetic fields.

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