The historical background of finding of electrogyration

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Abstract

190 years have passed from the experimental observations of optical activity by Arago and more than 30 years from the first observation of the spatial dispersion effect induced by electric field, which is now known as electrogyration. The aim of this article is to elucidate both the physical and historical fundamentals of discovery of the electrogyration effect and chronologize the main results concerned with the studies of electrogyration in the last thirty years.

Key words: electrogyration, optical activity, crystal optics.

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The problem of optical activity of crystals represents a quite complicated problem within the solid state physics. As a phenomenon of spatial dispersion (see [1–3]), it has come into notice of scientists and generated their intense discussions beginning from the first observations of optical activity in quartz crystals by D. Arago in 1811. Nonetheless, it has been only a simplest case of the optical activity, a natural one. Electrogyration, which consists in appearance (or changes) of the optical activity in the presence of external electric fields, has been recognized as a novel physical phenomenon only after a number of rather complicated experiments have been performed, accompanied with both successes and failures.

The difficulties mentioned above could be readily explained by the fact that the electrogyration represents a higher-order effect and, in many cases, it is concealed by the known electrooptic effect, the light polarization ellipticity, etc. As a consequence, small electrooptic effect or slight errors in the experimental geometry might, in principle, lead to the optical rotation, which are sometimes difficult to distinguish from the true electrogyration. A detailed analysis of the scientific and historical aspects of this problem, as well as the results obtained till 1983, may be found in the monograph [4] and the review [5]. However, the main attention in those works has been paid to that time's results on the electrogyration and the corresponding historical aspects, which have become somewhat outdated.

The changes in the optical activity sign induced by the external electric field have been observed for the first time in the ferroelectric crystals $LiH_3(SeO_4)_2$ by H. Futama and R. Pepinsky in 1962 [6], while switching enantiomorphous ferroelectric domains (the change in the point symmetry group of the crystal being $2/m \leftrightarrow m$). The observed phenomenon has been mistakenly explained as a consequence of specific domain structure (a replacement of optic axes occurred under the switching), rather than the electrogyration induced by spontaneous po-

larization. Hence, the problem of electrogyration has not been recognized at all at that time.

Two years later, in 1964, I. Zheludev [7] has predicted, on the basis of relations obtained in frame of tensor crystal optics, a possibility of appearance (or change) of the optical activity in crystals, which is proportional to the external electric field. The idea of optical activity induced by spontaneous polarization has been developed by K. Aizu [8] for the proper ferroelectrics, while L. Shuvalov and N. Ivanov [9] have discussed the problem of reversal of the optical activity occurred during the ferroelectric domain switching, following from a simple symmetry approach.

The first searches for the pure induced electrogyration effect performed in a few laboratories have not lead to positive results. For example, P. Lenzo et al. (see [10–12]) have mistakenly recognized the photorefraction effect in $Bi_{12}SiO_{20}$ crystals, which happens in the presence of external electric field, as an electrogyration. Considering the geometry of their experiments and the universal symmetry considerations, one can arrive at the conclusion that the electrogyration could not appear at all. Observation of the linear electrogyration in centrosymmetric $SrMoO_4$ crystals has been reported by Yu. Shaldin [13]. However, it has turned out to be erroneous, too: there the electrooptic effect has evidently been mistaken for the electrogyration. Later on (see [14]), the authors [13] have admitted their mistake. They have found the electrogyration in the mentioned crystals to be two orders of magnitude smaller than that reported in their previous experiments.

As a matter of fact, the first reliable results concerned with the observation of quadratic electrogyration effect in canonical quartz crystals have been obtained in 1969 by O. Vlokh [15, 16]. Those studies have been performed in the complicated conditions when disguising linear optical birefringence is present. Notice also that the term “electrogyration” has been used for the first time just in the works [15, 16]. Soon the linear electrogyration in α-HJO_3 (1971) [17] and $LiJ0_3$ (1974) [18] crystals has been revealed. Afterwards the results related to the linear electrogyration in the quartz crystals [19] have been subjected to partial revision and have been thoroughly explained using a general theory [20]. It is interesting to note in this respect that the Pockels effect (or the linear electrooptic effect) in the quartz crystals has also been observed later than its well-known quadratic analogue, the Kerr effect.

In its pure form (i.e., not interfered by the other crystal optical effects), the quadratic electrogyration has been observed for the first time in TeO_2 crystals by R. Vlokh et al. in 1989 [21]. Revealing, in 1975 [22], of quite large linear electrogyration effect in centrosymmetric $PbMo0_4$ crystals, where the natural optical activity is forbidden, has represented a principled and unambiguous experimental fact. The presence of the quadratic electrogyration in the quartz crystals has been confirmed by V. Shamburov and N. Romanova before long (1976) [23] and the linear electrogyration in $PbMo0_4$ has been repeatedly observed by M. Kostov et al. (1979) [24].

Extensive studies of the electrogyration in transparent dielectrics have rapidly increased a number of materials where the effect exists. For example, the works by H. Weber and S. Haussühl (1974–1983) [25-27] have been devoted to investigations of both the electrogyration and circular dichroism induced by electric field in alums. A number of studies of the optical activity in ferroic crystals has been performed by J. Kobayashi and Y. Uesu [28-33] and J. Etxebarria [34], using the so-called HAUP method. Finally, M. Kukhtarev et al. [35, 36] have investigated the influence of electrogyration on the recording of photorefractive grating in $Bi_{12}SiO_{20}$ crystals.

Regarding the optical activity changes occurred in the course of ferroelectric phase tran-

sitions, it should be noted that the experimental priority dates back to 1970 and belongs to the two research groups. One of them is that of K. Hermelbracht and H. Unruh [37], who have detected the effect in TGS crystals, though the authors have not connected it with the spontaneous polarization. The other group is that leaded by O. Vlokh. The latter group has obtained almost simultaneously the similar results for the same crystals [38, 39]. Analogously to electrooptics induced by the spontaneous polarization, this effect has been called as electrogyration induced by spontaneous polarization [38]. At the same time, O. Vlokh et al. have suggested its description on the basis of third-rank axial tensor associated with the paraelectric phase.

A number of important points in the studies of optical activity in ferroelectric crystal should be marked. These are the observations of the effect at the phase transition in $NaNO_3$ (M.-J. Chern and R. Phillips, 1972) [40] and $Pb_5Ge_30_{11}$ crystals (H. Iwasaki et al., 1971) [41, 42]. A further success has been concerned with revealing of exceptionally large induced electrogyration effect in the $Pb_5Ge_3O_{11}$ crystals and the solid solutions on their basis (O. Vlokh et al., 1977) [43]. The latter data have been confirmed afterwards by C. Konák et al. (1978) [44]. The authors [17] have predicted large electrogyration in the region of the Curie point, basing on the following considerations. It would be natural that, in the case of mechanically free (not clamped) crystals, the electrogyration effect should be determined by a contribution of piezogyration caused by piezoelectric deformation, i.e. it should possess a sharp, peak-like temperature dependence near the Curie point, which is known for the piezoelectric coefficients.

Thus, the electrogyration might be fruitfully used as a method for investigating ferroelectric phase transitions. One should mention in this relation the pioneer studies conducted by J. Kobayashi and Y. Uesu (1979) [45, 46], where the technique of quadratic electrogyration has been employed for investigations of pseudo-proper (improper, in their definition) phase transition in the KDP crystals. Later on, their results have been confirmed and further developed by O. Vlokh et al. (1985) [47, 48]

The obtained results have stimulated development of microscopic theory of the electrogyration based on the lattice dynamics approach. In particular, I. Stasyuk and S. Kotsur (1982) [49, 50] have used the method of Green's functions and obtained general expressions for the electrogyration tensor components, which are valid for the dielectric crystals of ionic type. Of course, the application of this approach does not mean that the phenomenological [19], thermodynamic [7] or oscillatory [51] models are restricted in some respects. Rather, it reveals a microscopic nature of this phenomenon in certain crystals.

Discovery of the electrogyration has given a pulse to studies of some new crystal optical effects, such as a piezogyration [52], magnetogyration [53, 54], acoustogyration diffraction of light [55], a gradient piezogyration [56], combined magneto-electrooptic activity [57], self-induced electrogyration [58] and a giant self-induced electrogyration in the exciton absorption region [59].

Finally, I would like to remind that the electrogyration might be treated as a first phenomenon of gradient nonlinear optics (see the general theoretical approach [60]). Indeed, from the standpoint of nonlinear electrodynamics, a gradient of the electric field of optical wave in the limits of unit crystalline cell would correspond to a macroscopic gradient of the external electrical field, if the field frequencies are properly transposed [60].

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