# SELF-INDUCED RESONANT OPTICAL ROTATION IN CRYSTALS KCl:Li

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The specific rotation of the resonant radiation polarization
plane predicted earlier for cubic crystals with tunneling
centers is observed in KCl containing $F_A$(Li) centers. The
rotation is supposed to be connected essentially with the
well-known off-center position of $Li^+$ ion. The simple the-
ory based on this assumption agrees with the experimental
data.

There are known many impurity cen-
ters in cubic crystals which have seve-
ral equivalent positions or orientati-
ons in the lattice cell [1,2]. The pro-
bability of the tunneling transition
from one position (orientation) to
another depends on the impurity elec-
tron state. As a rule this probability
in the excited state exceeds that in
the ground one. The excitation of the
impurity may be easily performed by re-
sonant radiation. The absorption coef-
ficient of a given center is anisotro-
pic because its local symmetry is lower
than cubic one. This anisotropy allows
to excite the impurities selectively
(depending on their orientation) by the
linearly polarized radiation. The fast
reorientation of the excited centers re-
sults in the optical alignment of the
impurities and hence in the anisotropy
of the impure cubic crystal as a whole.
In the theory of the optical orientati-
on [3] this anisotropy was taken into ac-
count self-consistently and was shown
to cause the turn of the orienting radi-
ation polarization plane to one of the

crystallographic axes. The nature of the
latter effect may be easily understood
if one consider the centers oriented
along the axes of the type $\langle 100 \rangle$ .

Let the radiation frequency lie in
the absorption band corresponding to the
excitation of the impurity by the radia-
tion with the electric vector $\vec{E}$ parallel
to the center axis. Then the light pro-
pagating in the $[00\bar{1}]$ direction (z-axis)
excites resonantly only the centers ori-
ented along x and y axes (the excitati-
on probability is proportional to $E_x^2$
and $E_y^2$ respectively). This leads to
the decrease in the total concentrati-
on of x- and y-centers (they go to z-
orientation) and hence to the decrease
in the resonant radiation absorption,
i.e. the self-transparency appears. The
latter effect becomes more pronounced
when the ratio of the reorientation ti-
me in the ground state to that in the
excited state grows because the transi-
tions from z-orientation take place in
the ground state only. If the radiation
is linearly polarized and $E_x > E_y$, then
x-centers are excited stronger than y-

centers. Hence the concentration of x-centers appears to be smaller than that of y-centers under stationary conditions and in its turn the x-component of the radiation is absorbed weaker than the y one. As a consequence the propagation of the resonant radiation is followed by the polarization plane rotation toward the x-axis.

To reveal the self-induced optical rotation experimentally the $F_A$(Li)-centers in KCl were chosen because of their high reorientation quantum efficiency [I]. In the "$F_A$ configuration", one nearest-neighbour cation of the F-center is replaced by the alkali ($Li^+$) ion. The impurity absorption spectrum consists of two bands. The $F_{AI}$ band corresponds to the transition polarized along the center axis, while the $F_{A2}$ band corresponds to the twofold degenerate transition polarized in the plane perpendicular to the center axis.

The $F_A$(Li)-centers in KCl were produced from F-centers, created by X-ray irradiation using the method, described in [I]. The optical orientation was performed by excitation of $F_{AI}$-transition with a He-Ne laser ($\lambda$=632,8 nm). To provide the angular polarization measurements the laser beam, passed through the Fresnel prism, traverses a path consisting of two Glan-Thompson polarizers, with the crystal, mounted in cryostat, between them. The experimental accuracy of the angle determination was $\pm I^\circ$.

The angle $\varphi$ between the $\vec{E}$-vectors of the incident and outgoing light versus the angle $\alpha$ between the $\vec{E}$-vector of the incident radiation and the $[I00]$-axis is plotted in Fig.I by points. It is seen that the polarization plane turns to the nearest between $[I00]$, $[0I0]$ directions. The sign of $\varphi$ is changed at $\alpha \approx 45^\circ$. In the range $0 \leqslant \alpha \leqslant 45^\circ$ (one-half period) the dependence $\varphi(\alpha)$ is somewhat asymmetrical and its maximum is shifted to to the nearest among $\langle I00 \rangle$ directions.
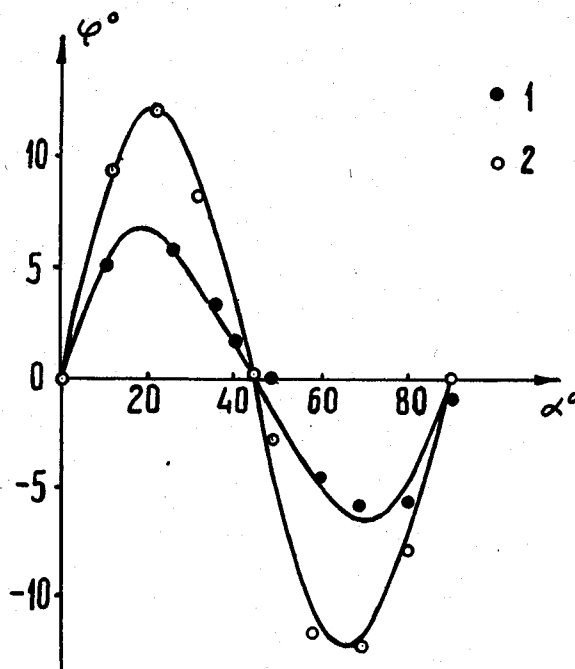


Fig.1.  Rotation of polarization for crystals with various $F_A$(Li) concentrations.  T=77$^\circ$K.  The X-ray irradiation doses are:  1- 7 Mrad; 2- 15 Mrad. The sample thickness is:  1- 0.26 cm; 2- 0.41 cm.

The rotation depends essentially on the $F_A$-concentration (radiation doze) and the thickness of specimen.

Although our experimental evidence agree qualitatively with the theoretical predictions [3] one cannot use [3] for the direct interpretation of these data. Indeed the ground state reorientation time for $F_A$(Li) centers in KCl is extremely large (it exceeds $I0^8$ sec at T< 200$^\circ$K [I] ). As a result the stationary polarization rotation is practically impossible for such centers in the model [3] . According to [3] in the configuration under consideration ($E_x, E_y \neq 0$; $E_z = 0$) all centers must go to the orientation $[00I]$ where they do not absorb the radiation. Hence after rather short time (it is inversely proportional to the light intensity and the excited state reorientation probability) the impurity absorption disappears and the polarization rotation vanishes.

The rotation we have observed may be connected with the off-center position of the $Li^+$ ion in the $F_A(Li)$ configuration discovered in [2]. The corresponding reduction of the site symmetry (as compared with $C_{4v}$ for the central $Li^+$ position) causes the mixing of the wave functions of the nondegenerate and degenerate excited states of the $F_A(Li)$ centers (their splitting is comparatively small and equals 0,26 ev). Consequently the polarization selection rules are softened and there is weak $F_{AI}$ absorption of the radiation polarized normally to the center axis. Then all centers appear to be excited by resonant radiation (with more or less probability depending on the polarization) and it is impossible to achieve a complete optical alignment. The latter was observed in [1,2] indeed.

To consider the optical orientation of the $F_A(Li)$ centers quantitatively it is convenient to generalize the approach [3] taking into account the change in the absorption selection rule. Neglecting the reorientation in the ground electronic state and taking into account the fast rotation of a $Li^+$ ion around the center axis the following kinetic equation may be obtained for the impurities:

$$\frac{d f_n}{dt} = -\sum_m \left( C_{nm} f_n - C_{mn} f_m \right) ;$$

$$\sum_n f_n = 1 ; \qquad n, m = x, y, z ; \qquad (I)$$

$$C_{nm} = A \left[ E_n^2 + \chi (E^2 - E_n^2) \right]; \quad E^2 = \sum_n E_n^2 .$$

Here $f_n$ determines the distribution of the centers over the orientations (optical equivalence of the orientations $[I00]$ and $[\bar{I}00]$ etc has been taken into account, cf. [3] ); $C_{nm}$ is the probability of the transition from n-th orientation to the m-th one; $E_n$ is the amplitude of the projection of the radiation electric vector on the n's axis. Parameter A presents the product of the resonant absorption cross section on the reorientation probability in the excited state ($I/4$

for $F_A(Li)$ in KCl [1] ). Parameter $\chi$ characterizes the ratio of the absorption cross sections for the light polarized normal and parallel to the center axis.

To describe the resonant radiation propagation in crystal self-consistently it is necessary to calculate the impurity contribution to the crystal polarization taking into account the center reorientation determined by Eq.(I). In the actual case of the linearly polarized radiation propagating in the direction $[00I]$ under stationary conditions $(t \gg C_{nm}^{-I})$ at $\chi \ll I$ the change of the angle $\Psi$ between the vector $\vec{E}$ and $[I00]$ may be shown to satisfy the equation

$$\frac{d\Psi}{dz} = -\frac{3}{2\ell_0} \frac{\sin 4\Psi}{2 + (\chi^{-I} + 2)\sin 2\Psi} ; \quad \Psi(0) = \alpha . \quad (2)$$

Here $\ell_0$ is the absorption constant in the absence of orientation ($\rho_x = \rho_y = \rho_z = I/3$). The radiation frequency is supposed to fall into the middle of $F_{AI}$ band and the contribution of impurities to the crystal refractive index is neglected. The corrections of the higher order in $\chi$ are neglected also. The angle $\varphi$ in Fig.I is connected with $\Psi$ by the obvious relation $\varphi = \alpha - \Psi(\ell)$ where $\ell$ is the crystal thickness. According to Eq.(2) the angle $\Psi$ is independent on the light intensity. This coincides with our observation (laser intensity was varied in 60 times).

The results of the numerical solution of Eq.(2) are plotted in Fig.I (solid curves) for $\chi = I/I5$. The curves I and 2 correspond to $I/\ell_0 = I$ and 2.2. The chosen value of $\chi$ is consistent in the order of magnitude with the ratio of intensities for $F_{AI}$ absorption bands before and after optical orientation which may be obtained from [1,2]. It seems to be reasonable also if one suppose that the shift of the $Li^+$ ion from the centre of the unit cell in $F_A(Li)$ system is of the same order as that of the isolated

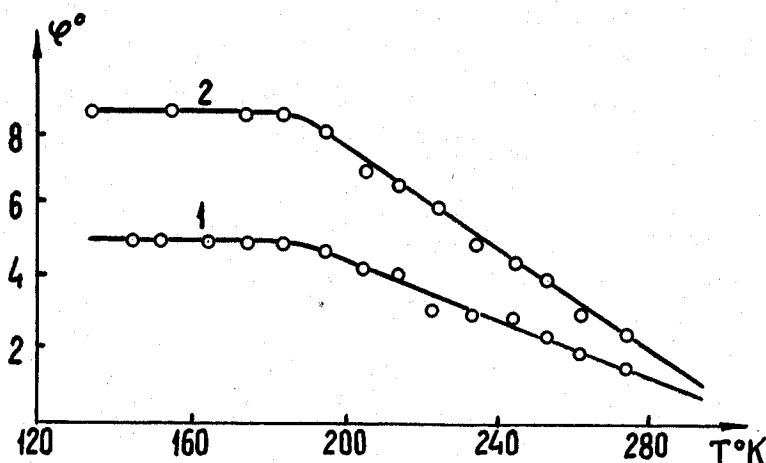Fig.2. Temperature dependence of the rotation
angle at α=30°. The curves 1 and 2 correspond to
the sample thicknesses 0.26 cm and 0.37 cm. X-ray
radiation doses are: 1- 7 Mrad; 2- 12 Mrad.

$Li^{\tau}$ in KCl (~0.2 lattice constant [4] ).
Our experimental error in the determi-
nation of $\chi$ makes up ~30%.

The temperature dependence of the
rotation is shown in Fig.2. It is seen
that $\varphi$ is independent on temperature up
to T 200°K. This fact can be explained
if one takes into account that the in-
crease in $l_o$ (according to Eq.(2) it re-
sults in the decrease in $\varphi$ ) is compen-
sated to some extent by the increase in
the parameter $\chi$ with temperature. At
T> 200°K $\varphi$ decreases gradually. This may

be caused by several reasons e.g. the de-
crease of the impurity absorption anisot-
ropy due to the overlap of $F_{AI}$ and $F_{A2}$
bands in this temperature range, expo-
nential growth of the ground state tran-
sition probability, the partial disso-
ciation of the $F_A$(Li) centers and so on.

It should be noted that an additi-
onal information concerning the origin
of the polarization selection rule vio-
lation may be obtained by the investi-
gation of the spectral dependence of the
optical rotation.

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