

METHODOLOGICAL NOTES

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Properties of acoustic polarization vectors in crystals and the phonon Hall effect

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Abstract. The properties of acoustic phonon polarization vectors in high-symmetry crystals are considered. Elliptic polarization of phonons in the presence of a magnetic field is shown to occur in crystals with the spin–phonon coupling.

Extensive literature is devoted to the theoretical and experimental study of vibrational spectra in crystals [1, 2]. In particular, the general properties of polarization fields and velocity branches of elastic waves in crystals have been discussed in papers on polarization crystal acoustics (see, for example, Refs [3–5]). Recently, however, an interesting new effect has been exposed [6], called the phonon Hall effect. The effect shows itself in solid insulators with a temperature drop, in which under the action of an external magnetic field an additional temperature gradient arises, the latter being normally directed to the initial heat flux and to the field. The effect was found to occur due to the phonon elliptical polarization induced by the magnetic field [7, 8]. Interest was drawn not only to the behavior of phonon polarization vectors (PPVs) in the magnetic field, but also to their general properties that are important in the heat conduction question. In the present paper, we consider a physical example of realizing acoustic activity (gyrotropy) in the presence of an external magnetic field. Perturbation of the Christoffel tensor in a solid insulator placed in a magnetic field is studied. An analogy is discussed of this effect with the mentioned much investigated acoustic phenomenon [9] and with even more popular optical gyrotropy [10].

In acoustic vibrations, the displacement vectors of atoms in an elementary cell oscillate synchronously, so that in what follows we shall consider a single-atom lattice without loss of generality. Acoustic vibrations of crystal can be expanded into orthonormalized lattice eigenmodes:

$$U_n^i = \sum_{ks} \sqrt{\frac{1}{2m\omega_{ks}N}}\, \exp\left(\mathrm{i}\mathbf{k}\mathbf{R}_n\right)\left(u_{ks}^i a_{ks} + u_{-ks}^{i*} a_{-ks}^{+}\right), \quad (1)$$

$$u_{ks}^{i*} u_{ks'}^i = \delta_{ss'}\,.$$

Here, ω_{ks} is the energy of the sth branch phonon, $\mathbf{k}$ is the wave vector, m is the atomic mass, N is the number of sites, $\mathbf{R}$ is the coordinate of the lattice site, and $\mathbf{u}_{ks}$ is the normalized PPV ($\hbar = 1$).

In the harmonic approximation, the polarization vector is a proper solution to the dispersion equation

$$\omega^2 u_{ks}^i = D_k^{ij} u_{ks}^j\,. \quad (2)$$

Actually, Eqn (2) is the set of three homogeneous linear equations whose eigenvectors are determined up to an arbitrary common phase. At different $\mathbf{k}$, these phases do not correlate. In particular, since expression (1) is explicitly Hermitian, the vectors $\mathbf{u}_{ks}$ and $\mathbf{u}_{-ks}$ are independent. Usually, the following condition (see Ref. [11]) is imposed:

$$\mathbf{u}_{ks} = \mathbf{u}_{-ks}^*\,. \quad (3)$$

However, we shall not do so. (Notice that with allowance made for anharmonism Eqn (2) becomes nonlocal and complicated constraints arise. At weak anharmonism these constraints are also weak and can be neglected.)

The dynamic real symmetrical matrix D_k^{ab} in the long-wave approximation represents the quadratic form

$$D_k^{ad} = \lambda^{abcd}\,\frac{k^b k^c}{\rho} = \frac{\Gamma^{ad} k^2}{\rho}\,, \quad (4)$$

where λ^{abcd} is the elastic modulus tensor, ρ is the density, and Γ^{ad} is the Christoffel tensor. The symmetry properties of tensor λ^{abcd} are specified comprehensively in Ref. [12] for all possible types of crystal symmetry. In particular, if the crystal lattice has a cube or square type symmetry, then for the coordinate axes chosen along the edges we have

$$D_k^{ab} = A_1 \delta^{ab} k^2 + A_2 \delta^{ab} k_a^2 + A_3 k_a k_b\,. \quad (5)$$

In the isotropic case, when $A_2 = 0$, the longitudinal mode energy is $\omega_{k\parallel} = k\sqrt{A_1 + A_3}$, and the transverse modes are degenerate ($\omega_{k\perp} = k\sqrt{A_1}$) and may have any PPVs that are orthogonal to the wave vector.

From formula (4) it follows that tensor D_k^{ab} and the eigenfrequencies ω_{ks} are invariants with respect to inversion. In this case, all the PPVs are odd functions of $\mathbf{k}$, or conversely, they are all even functions. According to Ref. [12], in high-symmetry crystals (cubic, rhombic, some tetragonal) the off-diagonal elements of the dynamic matrix are given by

$$D_k^{ab} \sim k_a k_b\,; \quad (6)$$

hence, they change sign when k_a or k_b changes sign — that is, in reflecting from the corresponding plane in the

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reciprocal space. In case (6), the frequencies do not change in reflection; however, the sign of the corresponding PPV component changes. To be sure of this fact, one may separate the off-diagonal terms out of dispersion equations (2):

$$(\omega_{ks}^2 - D_k^{aa}) e_{ks}^a = \sum_{b \neq a} D_k^{ab} e_{ks}^b \,, \tag{7}$$

where e_{ks}^a is the component of the normalized polarization vector without an external magnetic field. One can see that with the change of k_a sign the component e_{ks}^a should also change its sign, all the rest of the components keeping their signs. This property can be described by the formula

$$e_{ks}^a = \bar{e}_s^a(k) \operatorname{sign} k_a \,, \tag{8}$$

where the unit vector $\bar{e}_s^a(k)$ along with the frequency ω_{ks} are invariant with respect to reflection σ_a in the plane normal to the a-axis.

By using formula (8) we find that the vector product for polarizations of two modes changes sign in reflection:

$$(\mathbf{e}_s \times \mathbf{e}_{s'})^z = \operatorname{sign} k_x \operatorname{sign} k_y \, (\bar{\mathbf{e}}_s \times \bar{\mathbf{e}}_{s'})^z \,. \tag{9}$$

Let us consider the two-dimensional case in more detail [13]. A solution to Eqn (2) determines the dispersion law for two acoustic branches and, correspondingly, for two orthonormal PPVs:

$$(\omega_{ks}^2 - D_k^{xx}) \, e_{ks}^x = D_k^{xy} e_{ks}^y \,, \tag{10}$$

$$(\omega_{ks}^2 - D_k^{yy}) \, e_{ks}^y = D_k^{xy} e_{ks}^x \,. \tag{11}$$

It immediately follows that

$$\omega_s^2 = \frac{1}{2}(D_k^{xx} + D_k^{yy}) + s \, \frac{1}{2} \, R \,, \qquad s = \pm 1 \,, \tag{12}$$

$$R^2 = (D_k^{xx} - D_k^{yy})^2 + 4(D_k^{xy})^2 \,. \tag{13}$$

From here on, in obvious cases we omit the indication of dependence on $\mathbf{k}$ for the sake of brevity. In general, all three parameters D_k^{ab} are nonzero and not equal to each other. Hence, in two-dimensional (2D) crystals even on the edges with $D_k^{xy} = 0$ we have $R^2 \neq 0$ and the spectrum of acoustic phonons is nondegenerate. If condition (6) holds, then R^2 and $\omega_\pm^2$ are invariant with respect not only to inversion, but also to reflection from the edges: $(k^x, k^y) \to (k^x, -k^y)$ and $(k^x, k^y) \to (-k^x, k^y)$ (in a three-dimensional crystal this is reflection from the faces).

Let us find the polarization vectors. From equation (11) we obtain

$$e_s^x = (\omega_s^2 - D^{yy}) C_s \operatorname{sign} k^x \,, \quad e_s^y = |D^{xy}| C_s \operatorname{sign} k^y \,. \tag{14}$$

Normalization yields

$$C_s^{-2} = (\omega_s^2 - D^{yy})^2 + D^{xy\,2} = sR(\omega_s^2 - D^{yy})$$
$$= -sR(\omega_{-s}^2 - D^{xx}) > 0 \,.$$

Eigenvectors (14) and C_{ks} are determined accurate to the phase that arbitrarily depends on $\mathbf{k}$ and s. For definiteness, assume $C_{ks} = |C_{ks}|$. Then the normalized PPVs are real:

$$e_s^x = s \operatorname{sign} k^x \sqrt{\frac{s(\omega_s^2 - D^{yy})}{R}} \,,$$
$$e_s^y = \operatorname{sign} k^y \sqrt{\frac{s(\omega_s^2 - D^{xx})}{R}} \,, \tag{15}$$

and the following relations hold true:

$$(\mathbf{e}_+ \mathbf{e}_-) = 0 \,, \quad (\mathbf{e}_+ \times \mathbf{e}_-)^z = C_+ C_- D_k^{xy} R = \operatorname{sign} k^x \operatorname{sign} k^y \,. \tag{16}$$

PPV components (15) change sign both in inversion and in reflection, as they must in the case of a polar vector. In the course of slow rotation of $\mathbf{k}$, the PPV components do not turn to zero and exhibit a stepwise change as vector $\mathbf{k}$ crosses the corresponding axis. In the process, one of the phonon polarization vectors is longitudinal, and the other is transverse in the isotropic model only. It is shown [3, 4] that the stepwise change can be eliminated by corresponding redefinition of the polarization vectors. However, the change is important in the thermal conductivity problem in which the integral is taken over all directions of the wave vector, and such a redefinition would be inconvenient [see expression (36) below].

Notice that vector (15) does not satisfy constraint (3). Nevertheless, constraint (3) holds true if vector (15) is multiplied by an imaginary unit and PPV may be considered either real or Hermitian and purely imaginary. If PPVs are taken as real odd functions of $\mathbf{k}$, then expansion (1) takes the form

$$U_n^i = \sum_{ks} \sqrt{\frac{1}{2m\omega_{ks}N}} \exp(\mathrm{i}\mathbf{k}\mathbf{R}_n) \, e_{ks}^i (a_{ks} - a_{-ks}^+) \,. \tag{17}$$

Consider the origin of phonon elliptical polarization in the case of a Hermitian dynamic matrix with complex terms. Let the crystal insulator comprise paramagnetic particles (atoms or molecules) magnetized by an external magnetic field. The so-called spin-phonon interaction (SPI) of these particles with acoustic vibrations of the lattice results in renormalization of phonons. The Hamiltonian with SPI included takes the following form [7, 8]

$$H = \sum_n \frac{\mathbf{P}_n^2}{2m} + \frac{m}{2} \sum_{nn'} D_{nn'}^{ab} U_n^a U_{n'}^b + g \sum_n \big(\mathbf{M}_n, [\mathbf{U}_n \times \mathbf{P}_n]\big) \,. \tag{18}$$

Here, $\mathbf{U}_n$, $\mathbf{P}_n$, and $\mathbf{M}_n$ are, respectively, the displacement vector, momentum, and magnetic moment of the particle at site n. The magnitude of interaction energy g is determined by a crystal field [14, 15]. In describing long-wave vibrations of a lattice, the magnetic moment at a site can be replaced by its average value $\mathbf{M}$, which is proportional to the magnetic field strength. From formula (18) we obtain the equations of motion:

$$\frac{\mathrm{d}U_n^a}{\mathrm{d}t} = V_n^a = \frac{\partial H}{\partial P_n^a} = \frac{P_n^a}{m} + e_{abc} \, g M^b U_n^c \,, \tag{19}$$

$$\frac{\mathrm{d}P_n^a}{\mathrm{d}t} = -\frac{\partial H}{\partial U_n^a} = -\sum_{n'} m D_{nn'}^{ab} U_{n'}^b + e_{abc} \, g M^b P_n^c \,, \tag{20}$$

where e_{abc} is the third-rank unit tensor that is antisymmetric with respect to all three indices, and $e_{123} = 1$.

It is important that SPI affects the relationship between the velocity and momentum of the particle [see Eqn (19)]. In metals, a similar relationship that holds true for an electron in the field of a magnetic atom is associated with the Berry phase. However, there is a noticeable difference. In metals, this relationship entails a curling of the electron trajectory, which results in the anomalous Hall effect [16–19]. According to this equation, the character of particle oscillations in insulators changes, but the phonon motion remains rectilinear, though with changed polarization.

Hereinafter, we make allowance for SPI in the linear approximation. Then, in the last term in expression (20) we may make the substitution $P_n^c \to mV_n^c$ and obtain the equation of vibration:

$$\ddot{U}_n^a = \sum_{n'} D_{nn'}^{ab}(U_n^b - U_{n'}^b) + 2e_{abc}\, gM^b \dot{U}_n^c \,. \tag{21}$$

In the momentum representation, equation (21) takes the form

$$\omega_{ks}^2 u_{ks}^a = \tilde{D}_k^{ab} u_{ks}^b \,, \tag{22}$$

where the dynamic matrix is now perturbed by the external magnetic field:

$$\tilde{D}_k^{ab} = D_k^{ab} - \mathrm{i}e_{abc}G^c \,, \tag{23}$$

with $\mathbf{G} = 2\omega g\mathbf{M}$, $(\mathbf{u}_{ks}^* \mathbf{u}_{ks'}) = \delta_{ss'}$. It should be noted that the imaginary part of $\tilde{D}_k^{ab}$ is the antisymmetric tensor which is odd with respect to $\mathbf{G}$. In the zero order with respect to SPI, Eqn (22) is equivalent to Eqn (2).

We shall solve equation (22) in the linear approximation in SPI. We seek the renormalized PPV in the form of a small addition to the zero approximation: $u_s^a = e_s^a + \delta e_s^a$. Let us rewrite equation (22) in the form

$$(\omega_s^2 + 2\omega_s \delta\omega_s)\,(e_s^a + \delta e_s^a) = D^{ab}(e_s^b + \delta e_s^b) - \mathrm{i}(e_{abc}G^c)e_s^b \,. \tag{24}$$

The real part of this equation yields $\delta\omega_s = 0$ — that is, the spectrum and group velocity of phonons $\mathbf{c}_{ks} = \partial\omega_{ks}/\partial\mathbf{k}$ in the linear (with respect to SPI) approximation are not renormalized. The imaginary part of equation (24) determines renormalization of the polarization vector:

$$(\omega_s^2 \delta^{ab} - D^{ab})\delta e_s^b = -\mathrm{i}(e_{abc}G^c)e_s^b \,.$$

The solution to this equation is as follows:

$$\delta e_s^b = \mathrm{i} \sum_{s'(s' \neq s)} K_{ss'} e_{s'}^b \,, \tag{25}$$

and also

$$K_{ss'}(k) = K_{s's}(k) = (\omega_{ks}^2 - \omega_{ks'}^2)^{-1}[\mathbf{e}_{ks} \times \mathbf{e}_{ks'}]\,\mathbf{G} \,, \qquad s' \neq s \,. \tag{26}$$

Thus, the inclusion of SPI results in the real vector $\mathbf{e}_{ks}$ acquiring an orthogonal imaginary addition — that is, it becomes elliptically polarized.

From expression (26) it follows that the renormalization of PPV increases as the spectrum of two adjacent modes is close to degeneration. Therefore, it is interesting to study the problem in the isotropic body model [20], in which case the transverse modes ω_0 are degenerate for all directions of $\mathbf{k}$. Equation (22) in this model takes the form

$$We^a = \lambda \hat{k}^a(\hat{\mathbf{k}}\mathbf{e}) - \mathrm{i}[\mathbf{e} \times \mathbf{G}]^a \,. \tag{27}$$

Here, $W = \omega^2 - \omega_0^2$, $\lambda = wk^2$, where w is the difference of squares of longitudinal and transverse velocities of sound without a magnetic field, and

$$\hat{\mathbf{k}} = \frac{\mathbf{k}}{k} = (\sin\theta\cos\varphi,\ \sin\theta\sin\varphi,\ \cos\theta) \,,$$

$\mathbf{G} = (0,\ 0,\ G)$, $\mathbf{G}\hat{\mathbf{k}} = G\cos\theta$, $Q = G\sin\theta$. Let us introduce normalized unit vectors:

(1) $\hat{\mathbf{k}}$,

(2) $\hat{\mathbf{m}} = \dfrac{(\mathbf{G} - (\mathbf{G}\hat{\mathbf{k}})\hat{\mathbf{k}})}{Q} = (-\cos\theta\cos\varphi,\ -\cos\theta\sin\varphi,\ \sin\theta)$,

(3) $\hat{\mathbf{n}} = \hat{\mathbf{k}} \times \hat{\mathbf{m}} = Q^{-1}\,\hat{\mathbf{k}} \times \mathbf{G} = (\sin\varphi,\ -\cos\varphi, 0)$.

The components of vector $\mathbf{e}$ can be found from equation (27):

$$\mathbf{e} = \xi\left(\frac{Q}{W - \lambda}\,\hat{\mathbf{k}} - \frac{\mathbf{G}\hat{\mathbf{k}}}{W}\,\hat{\mathbf{m}} - \mathrm{i}\hat{\mathbf{n}}\right), \tag{28}$$

and we arrive at the equation for eigenvalues:

$$\frac{Q^2 W}{\lambda - W} + W^2 - (\mathbf{G}\hat{\mathbf{k}})^2 = 0 \,. \tag{29}$$

Dispersion equation (22) determines the polarization vectors accurate up to the phase. Assuming the phase of parameter ξ is zero, the modulus of ξ is given by the normalization

$$\xi^{-2} = \left(\frac{Q}{\lambda - W}\right)^2 + \left(\frac{\mathbf{G}\hat{\mathbf{k}}}{W}\right)^2 + 1 \,. \tag{30}$$

In inversion $\hat{\mathbf{k}}, \hat{\mathbf{m}}, \hat{\mathbf{n}} \to -\hat{\mathbf{k}}, -\hat{\mathbf{m}}, -\hat{\mathbf{n}}$, the polarization vector (28) changes sign as it is the polar vector. The complexity of vector (28) means that it is elliptically polarized.

Formulas (28)–(30) are valid for all G. Let us next assume the interaction of phonons with internal degrees of freedom of ions (molecules) is weak, $G \ll \lambda$. In this case, from equation (29) we obtain the single longitudinal mode: $W_\parallel = \lambda$, $\mathbf{e} = \hat{\mathbf{k}}$.

In the zero approximation, the parameter W for transverse modes is zero, and with G taken into account, the quantity

$$W_\eta = \frac{1}{2}\left(-\frac{Q^2}{\lambda} + \eta\sqrt{\left(\frac{Q^2}{\lambda}\right)^2 + 4(\mathbf{G}\hat{\mathbf{k}})^2}\,\right), \quad \eta = \pm 1 \tag{31}$$

characterizes their splitting:

$$\Delta = \omega_+ - \omega_- = \frac{1}{\omega_0}\sqrt{\left(\frac{Q^2}{2\lambda}\right)^2 + (\mathbf{G}\hat{\mathbf{k}})^2} \,. \tag{32}$$

The splitting is minimal near the equator [$|\cos\theta| < G/\lambda$, $\Delta = Q^2/(2\lambda\omega_0)$]. For all the remaining directions of $\hat{\mathbf{k}}$ we

have

$$W_\eta = \eta|\mathbf{G}\hat{\mathbf{k}}|\,, \qquad \Delta = \frac{G\,|\cos\theta|}{\omega_0}\,. \tag{33}$$

In the zero approximation, the real and imaginary parts of each $\mathbf{e}_\eta$ are orthogonal and equal in magnitude:

$$\mathbf{e}_\eta \approx -\frac{1}{\sqrt{2}}\left[\eta(\text{sign }\cos\theta)\,\hat{\mathbf{m}} + \mathrm{i}\hat{\mathbf{n}}\right]. \tag{34}$$

This means that at infinitesimal (but nonzero) $\mathbf{G}$ the transverse phonons have just circular polarization [21].

The direction of the polarization vector (34) changes stepwise while crossing the equator. But if we alter the numbering of transverse modes and discard the modulus in expression (33), $W_\eta = \eta\mathbf{G}\hat{\mathbf{k}}$, the projection of vector (34) onto the $\hat{\mathbf{m}}$-axis becomes constant. This is always the case when levels are crossed.

In the linear approximation with respect to SPI we find the deviation from transversality:

$$\mathbf{e}_\eta\hat{\mathbf{k}} = -\frac{\xi_\eta Q}{\lambda}\left(1 + \frac{W_\eta}{\lambda}\right). \tag{35}$$

In the result of solving the heat conduction problem in solid insulators at low temperatures [7, 8] (when only acoustic branches of vibrations are excited), it was shown that in the coordinate system in which the magnetic field and magnetic moment are directed along the z-axis and the temperature gradient ∇T is directed along the x-axis, the transverse component of the heat conductivity tensor is expressed in the form

$$\varkappa_{12}^{yx} \approx \frac{2}{V}\sum_k K_{12}(\mathbf{k})\omega_{k1}\omega_{k2}(c_{k2}^y - c_{k1}^y)\,\mathrm{Re}\,A_{12}^x(\mathbf{k})\,, \tag{36}$$

where the function $K_{12}(\mathbf{k})$ is determined according to formula (26) and the density matrix in the zero approximation in SPI has the form

$$A_{pq}^x(\mathbf{k}) = \frac{F_{ss'}(\mathbf{k})\,c_s^x(\mathbf{k}) + F_{s's}(\mathbf{k})\,c_{s'}^x(\mathbf{k})}{\omega_{ks} - \omega_{ks'}}\,,$$
$$F_{ss'}(\mathbf{k}) = -\frac{\omega_{ks}\Omega_{pq}}{2T^2\Omega_{pp}}\,N_{ks}(1 + N_{ks})\,, \tag{37}$$

where $p = \mathbf{k}s$, $q = \mathbf{k}s'$, and Ω_{pq}, Ω_{pp} are the phonon collision frequencies.

One can see from formula (26) that the polarization vectors of acoustic modes play a noticeable role in the formation of the effect under study. The factor $K_{12}(\mathbf{k})$ in expression (36) arises due to the presence of elliptical polarization, and the integral taken over the directions of the wave vector is nonzero due to the step character of vector product (9) which is included in function $K_{12}(\mathbf{k})$.

In experiment [4], the effect was observed in the $Tb_3Ga_5O_{12}$ terbium–gallium garnet crystal at a temperature of 5.45 K in the presence of an initial temperature gradient and magnetic field normal to it. The transverse heat flux arises due to a combination of two significant factors. First, it is the elliptical polarization that arises due to the spin–phonon interaction between phonons and paramagnetic ions whose multiplets are split by the crystal field. Second, the effect is connected with the fact that under the action of the temperature gradient the correlated motion of two phonon modes arises with the emergence of off-diagonal (with respect to modes) elements of the density matrix $A_{pq}^x(\mathbf{k})$. Notice that the collision frequencies enter into expression (36) only in the form of a ratio, because the phenomenon develops in two stages: first, the temperature nonuniformity causes the nonequilibrium distribution of phonons $f_p \sim 1/\Omega_{pp}$; and only after that the correlator $\rho_{pq} \sim \Omega_{pq}$ arises against the background of this distribution, which forms the transverse heat flux.

Finally, also noteworthy is the suggesting itself analogy of the phenomenon considered with the widely studied gyrotropy phenomenon in acoustics [9] and with more widely known optical gyrotropy [10].

It is appropriate that the discovery of the phonon Hall effect was initiated by the other effect connected with the influence of magnetic field on sound propagation in matter (it is termed the acoustic Faraday effect by analogy with the magneto-optical Faraday effect), which shows itself as rotation of the polarization plane of shear waves [22]. Notice that the photon Hall effect [23], which was discovered a short time before the phonon Hall effect, also derives from the magneto-optical Faraday effect: the difference in the propagation velocities for two circularly polarized waves results in a turn of directional pattern of Rayleigh rotation, which causes a transverse flux of photons in their numerous scattering on defects. Similarly, the acoustic Faraday effect is an indirect indication of the existence of the phonon Hall effect.

The effects mentioned are in fact various manifestations of acoustic or optical activity (gyrotropy). This explains the similarity of the equations derived in the present work with those from paper [10], where the degeneration of isonormal electromagnetic waves was considered. For example, taking into account gyrotropy transforms the permittivity tensor to the complex Hermitian one, and the magnetic field similarly perturbs the Christoffel tensor in solid insulators. However, in contrast to the natural optical activity of crystals considered in paper [10], anisotropy may be induced — that is, may arise in optically (acoustically) isotropic media under the action of an external factor changing the local symmetry. In our case, such action is implemented by the magnetic field; hence, from the viewpoint of gyrotropy manifestation, the acoustic Faraday effect is the most similar to the phonon Hall effect among the phenomena mentioned.

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