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# Spin angular momentum induced by optical quasi-phonons activated in birefringent uniaxial crystals

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A B S T R A C T

The present report formally establishes the expression of the angular momentum of the quasi-phonons induced by linearly polarized light. The transferred mechanical torque due to phonons is then determined from the spin angular momentum and is shown to be measurable from Raman scattering experiments. To investigate this, the electric field due the excited dipoles and the associated macroscopic dielectric polarization vectors were first calculated using a lattice dynamical model in order to derive in a second step the analytical expression of the angular momentum density arising from the inelastic light scattering by quasi-phonons. The numerical results of the calculated angle dependent mode electric fields and the induced spin angular moments as well as the transferred torques were analyzed with regard to some typical behaviors of the interacting modes and it is shown that the fluctuations of the effective charges is their main origin.

## 1. Introduction

The concept of the non-invasive rotation of the dielectric materials of small size has been the subject of intense investigations mainly in the last three decades. The pioneering work of Beth in 1936 [1] has known a revival of interest with the advent of optical manipulation of nano-sized object [2,3] and also with the decisive work of Allen [4]. Micron-sized materials are often displaced either by optical forces or rotated by optical torques. There are two main origins of the optical torque, that is the torque due to charges and the torque caused by the exciting field [4–7]. These torques result mainly from spin angular momentum (SAM) and orbital angular momentum (OAM) transfers. However the splitting of the light induced angular momentum, subject of many studies [8], into SAM and OAM and also the conversion of SAM to OAM is currently investigated and still remains of great interest [8,9]. An excellent review of these topics is published by Marrucci [8]. Experimentally, mechanical torque induced by linearly polarized light is determined by the torsional balance measurements whose sensitivity can reach $10^{-17}$ Nm subject to certain constraints [10,11]. Torque is currently theoretically studied as either arising from the quantum fluctuations [10,11], that is the Casimir torque or the scattering of light in which case the birefringent scatterers are considered as wave plates thus modifying the polarization state of the light [1,12,13]. Nevertheless when a linearly polarized electromagnetic radiation is incident on a birefringent dielectric homogeneous medium, one may also expect there to occur an inelastic scattering of the incident wave (Brillouin or Raman scattering). As will be seen in the present paper, this type of interaction of light with matter can induce, as with elastic scattering [13], another angle dependent phonon angular momentum and transferred torque.

The main aim of the present paper is to derive self-consistently the analytical expression of the angular momentum density and that of the spin angular momentum induced by the interaction of the optical polar phonons in $LiNbO_3$ (LN) and in α-quartz uniaxial crystals. To satisfy such objectives, the macroscopic electric fields associated with the angle dependent normal mode coordinates has been first analytically determined as well as the dielectric polarization and then compared with those numerically calculated by [14]. From the cross product of these fields, we show in the second step, how the angular momentum density is related to the electric displacement and the absolute walk-off angle. By connecting the quasi-normal coordinates to the imaginary part of the response function, the expression of the angular momentum of the quasi-phonons and that of the transferred torque are finally established. The paper is structured as follows: Section 2 is devoted to the theoretical derivation of the angular momentum of quasi-phonons while Section 3 focuses on the numerical results for different values of the angular rotation position of the studied crystal then followed by discussion in Section 4.

## 2. Theoretical approach

The angular momentum density (AMD) induced by a polarized light

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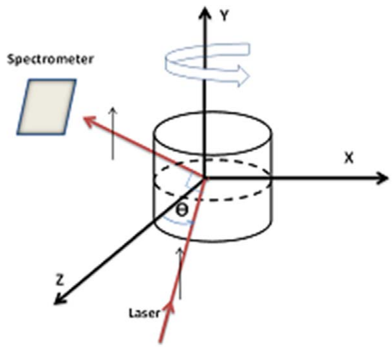


**Fig. 1.** Proposed Raman experimental ninety degree scheme according to the geometrical configuration pz+qx(yy)-qz+px with p and q changing from 0 to 1 that corresponds to the rotation of the LN crystal around the y-dielectric axis with parallel polarization filters (yy). It would be assumed that the crystal is suspended along the y-axis. The rod-shaped thickness is exaggerated to show the scattering plane e.g., (zx). In this geometry of scattering, neither the phonon wave vector nor its polarization is parallel to the dielectrics axes. Such oblique phonon wave vector or fundamental phonon with oblique polarization state is recognized as quasi-phonon.

wave impinging on a birefringent crystal can be arisen from the elastic light scattering, the quasi-elastic and the inelastic light scattering. We restrict ourselves in the present study to the part of the angular momentum density originated from the inelastic light scattering by interacting polar optical phonons, which can be written according to [20]:

$$\overrightarrow{K} = \overrightarrow{P} \times \overrightarrow{E}, \tag{1}$$

where $\overrightarrow{K}$ is the induced AMD per chemical formula unit due to the interacting phonons, $\overrightarrow{P}$ and $\overrightarrow{E}$ are the induced macroscopic dielectric polarization and the mode electric field respectively at the sublattice site. The calculation of the total phonon AMD needs to take into account the number of the formula unit or the sublattice per unit cell. To avoid confusion, in (1) only the electric field due to the excited dipoles is considered in the present study and not the field associated with the light wave impinging the crystal. We simply assumed a linearly polarized plane wave satisfying the geometrical configuration as displayed by Fig. 1, thus allowing for the observation of mixed or oblique Raman modes called quasi-phonons according to the analogy with quasi-normal modes termed by [14] with the view to pointing out only the part of the optical torque related to the electric dipoles [5,7]. Our next concern is the derivations of the dielectric polarization and the mode electric field.

### 2.1. Formulation of the quasi-normal coordinates, of the macroscopic electric field and the induced polarization vectors

The angular behavior of the quasi-phonons is well studied both experimentally and by the lattice dynamical model computed by [15]. A second approach [16] based on the quantum mechanical considerations have been applied to result in the angular dispersion of quasi-phonons frequencies in the LN crystal. In spite of the apparent age of the model of [15], it presents some advantages that rely among others on the analysis of the Raman intensity, hence the Raman polarizability. Another feature which motivates the selection of the approach of Ref [15] is the opportunity to establish analytically the expressions of the mode electric field and macroscopic polarization of the interacting modes.

To determine the electric field corresponding to the quasi-normal mode, one might first solve the eigenvalue problem. Indeed, the angle dependent frequencies of the quasi-phonons and the corresponding quasi-normal coordinates can be obtained by solving the following set of secular equations:

$$\left( (\omega_j^2 - \omega^2)\delta_{jj'} + \frac{4\pi}{\varepsilon^{\infty}(\hat{q})}[\hat{q}.\overrightarrow{M}(j)][\hat{q}.\overrightarrow{M}(j')] \right) u_{j'} = 0 \tag{2}$$

where $\omega_j$ is the j$^{\text{th}}$ fundamental mode frequency, $u_j$ is the angle dependent normalized eigenvector component or the dimensionless quasi-normal coordinate, $\hat{q}$ is the unit vector of the phonon wave vector and $\varepsilon^{\infty}(\hat{q})$ is the screening factor of the high frequency dielectric response with:

$$\varepsilon^{\infty}(\hat{q}) = \varepsilon_x^{\infty} q_x^2 + \varepsilon_y^{\infty} q_y^2 + \varepsilon_z^{\infty} q_z^2. \tag{3}$$

For the case of the uniaxial crystal point groups, (3) reduces to:

$$\varepsilon^{\infty}(\hat{q}) = \varepsilon_{A_1}^{\infty} \cos^2\theta + \varepsilon_E^{\infty} \sin^2\theta. \tag{4}$$

where $\theta$ is the angle between the phonon wave vector and the z axis here considered for the $C_{3v}$ point group as the optical axis of the uniaxial crystal and the component of the vector transformation of the $A_1$ irreducible representation while the x axis refers to the doubly degenerated E irreducible representation polar direction and finally $\overrightarrow{M}(j)$, the complex-valued dielectric polarization along one principal dielectric axis, can be considered according to [15] as scalar number with arbitrarily fixed zero phase and calculated from the oscillator strength (Eq. (A8)). Because of further developments based on the considered model we recall in the Appendix A some relevant features of the work of [15] which will be needed hereafter.

Indeed, according to [15], the mode electric field can calculated by combining relations (A2), (A5) and (A7) from the Appendix A thus allowing to establish relation (A8) in its final form that is,

$$\overrightarrow{E} = \frac{-4\pi}{\varepsilon^{\infty}(\hat{q})} \sum_j u_j(\lambda)[\hat{q}\overrightarrow{M}(j)]\hat{q}. \tag{5}$$

As mentioned by [14], it appears that the electric field vector is antiparallel to the phonon wave vector $\hat{q}$. In the context of the LN crystal, relation (5) can be written:

$$\overrightarrow{E_{LN}} = \frac{4\pi N_u}{\varepsilon^{\infty}(\hat{q})}\left(\hat{q}_z \sum_{j=1}^{4} u_j(\lambda) M(j) \cos\theta + \hat{q}_x \sum_{j=1}^{9} u_j(\lambda) M(j) \sin\theta\right), \tag{6}$$

where $q_j$ (j = z,x) are the unit vectors parallel to the polar direction of the $A_1$ and E symmetry species, $N_u$ is the number of sublattices per unit cell volume which is 8 for the LN crystal, λ stands for the eigenvalue and $E_{LN}$ is the macroscopic electric field. Fig. 2 illustrates the specific use of indices j, λ. Note that (5) and (6) are expressed per length unit since the quasi-normal coordinates are normalized. Relation (6) is slightly different to that given by Hwang [17] by the screening dielectric factor and also indicates that the macroscopic electric field is modulated by the eigenvectors thus meaning the behavior of the absolute

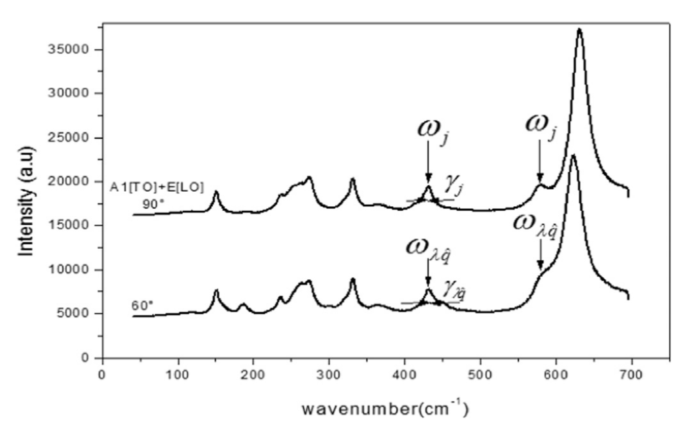


**Fig. 2.** Illustrated Raman spectra recorded at 300 K for LN under the ninety degree scheme for the angular rotation of the crystal corresponding to 60° and 90°; index j is assigned to the fundamental $A_1$ and E modes while $\lambda\hat{q}$ refers to the quasi-modes; both indices addressed to frequency and damping of a Raman scattered line.