1 The scattering chamber

The scattering chamber of BEAR has been used for the first time for the present measurements. Therefore part of the work reported in this thesis was devoted to its setup.

The chamber features characteristics well beyond the requirements of this work: it shows original aspects mainly to be exploited in the field of XAS and light scattering according to the mission of BEAR. Consequently a detailed description is presented, illustrating also those distinctive features not directly related to the present work of thesis.

The experimental chamber provides the possibility of measuring both photons and electrons: it is equipped with three light detector (photodiodes) and one electron analyzer. The electron analyzer, and the photodiode fixed on it, can move with two degrees of freedom, which, combined together, allow it to be positioned in any point of the hemisphere above the sample. A picture of the detector with the sample holder and the manipulator is shown in Figure 1

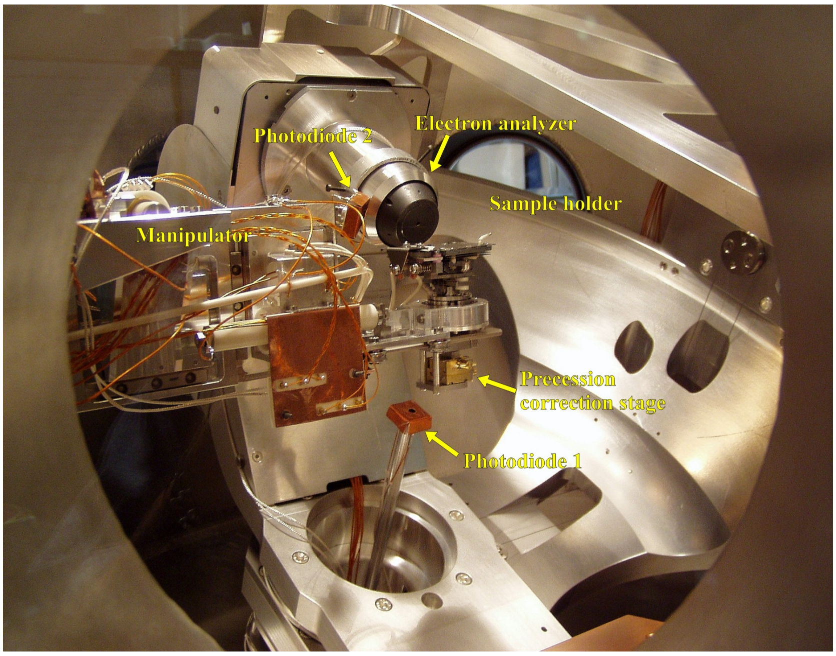

Figure 1: Light and electron detector of the experimental chamber. It is visible also the manipulator and the sample holder. The beam comes from the right side of the picture.

Five different reference systems are defined:

- Ω_L is the system fixed in the laboratory. The positions are expressed in terms of the three Cartesian coordinates x_L, y_L, and z_L, choosing x_L parallel to the beam axis with positive direction the direction of propagation of the light beam, z_L vertical pointing upwards, and y_L according with the "right hand rule".

- Ω_C is fixed with the chamber, and can rotate, with respect to Ω_L around the beam axis in order to obtain the changing of the polarization of the light impinging the sample. The angle describing the position of the chamber is called Ψ_C and is defined 0 when the three axes x_C, y_C, and z_C are parallel to x_L, y_L, z_L respectively.

- Ω_M is fixed with the sample holder and is defined having always the y_M axis parallel to y_C axis around which it can rotate describing an angle θ_M. θ_M is strictly related to incidence angle of the light on the sample ($\theta_M = \pi/2 - \theta_{inc}$, being θ_{inc} the incidence angle). θ_M is zero when the x_M axis is parallel to x_C and x_L. The sample can be also rotated around the z_M axis (azimuthal angle ϕ_M) to be oriented in specific directions with respect to the incident light. The translational movements of the sample (x_M, y_M, and $z - M$) and the azimuthal angle ϕ_M are driven in vacuum by piezo-inchworm motors (Burleigh, type UHVL-025) (see Figure 3). Manipulator positions are read directly in vacuum by absolute measurement systems, made by A.P.E. Research, Trieste) based on position sensitive detector (PSD) technology (see Figure 3).

- Ω_A is fixed with the detector (electron analyzer and/or photodiode); the y_A axis is parallel to y_M and y_C. The position of the detector is described, rather than by x_A, y_A, or z_A, by two angles: θ_A (polar angle) is the angle describing the rotation of the analyzer shaft around the y_A axis, ϕ_A (azimuthal angle) is the rotation of the detector around an z_A axis, fixed on the shaft. $\theta_A = 0$ when the z_A axis is parallel to z_C, $\phi_A = 0$ when the electron analyzer is parallel to the y_A axis and points toward the positive direction.

- Ω_S is a frame of reference fixed on the sample. It can be translated in space and the axis z_S, normal to the sample surface, can change orientation, in order to become parallel to the z_M axis (x_S and y_S will accordingly change their direction). This movement, controlled by means of a vacuum compatible screw driver, is useful to correct systematic errors deriving from sample normal misalignments with the scattering plane. Once made this corrections, the Ω_S frame of reference is considered indistinguishable from the Ω_M system.

These five reference systems are represented in Figure 2.

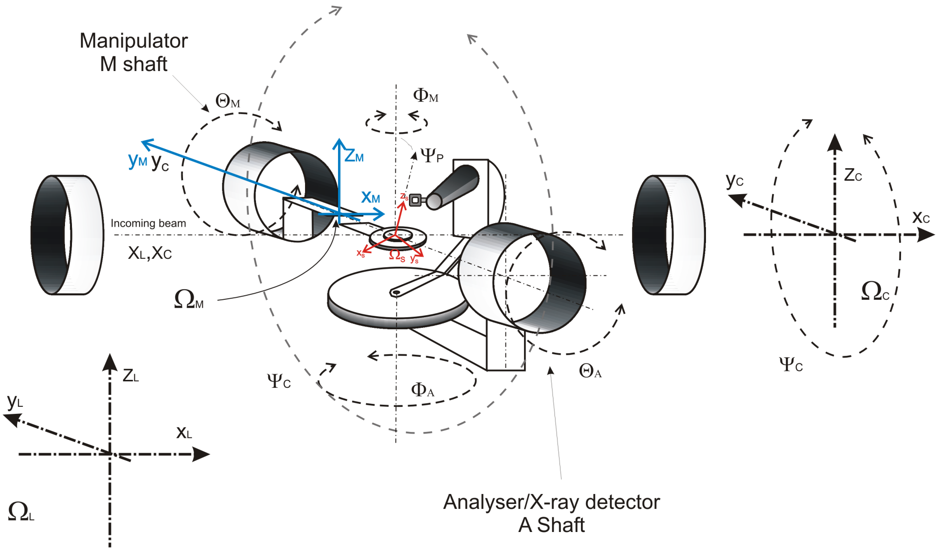

Figure 2: Definition of the frame of reference of the scattering chamber and relative movable bodies including sample, manipulator's shaft, detector's shaft, electron analyzer, and photon detector.

The movements of the analyzer (θ_A and ϕ_A), the polar angle of the sample (θ_M) and the rotation of the whole chamber (angle Ψ_C) are driven by stepper motors controlled by a computer.

Instead of thinking the experimental geometries referred to the frame of reference of the chamber (Ω_C) or of the laboratory (Ω_L), it is generally better to refer the geometrical parameters to the system of reference of the sample. In this case the sample is fixed with the observer and the orientation of the sample, the position of the detector, the direction and polarization of the incoming light are described accordingly to the sample. We aim to find the relations which connects these parameters, expressed in the Ω_M system of reference, with those parameter directly controlled by the computer.

We need to define three vectors:

1. a vector fixed with the sample, not parallel to its normal, whose projection on the X_M-Y_M plane defines the *sample azimuth* ϕ_M; this vector is useful to correlate some particular feature of the sample (for instance a particular cristallographic direction, some characteristic of the sample, etc.) to the Ω_M reference system.

2. the wave vector $\vec{k}$ of the incident electromagnetic field, and with it the

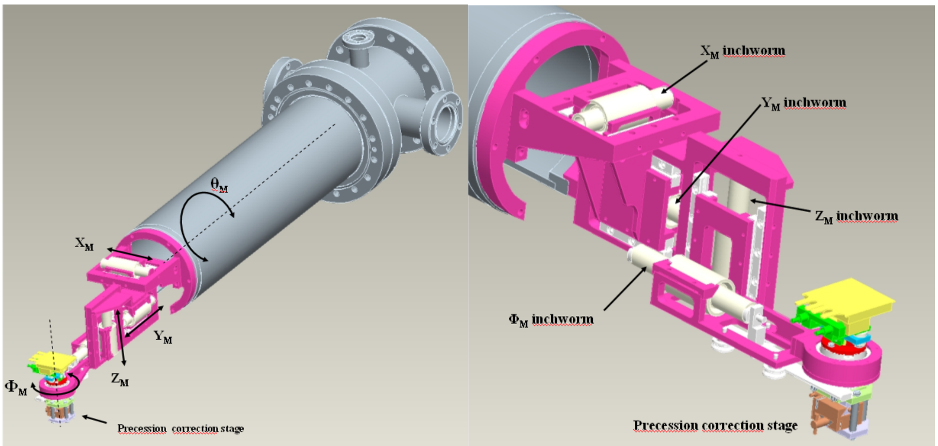

Figure 3: The manipulator of the experimental chamber: on the left are indicated the degrees of freedom and, on the right, the inchworm motors that drive the manipulator movements.

directions of the horizontal and vertical components of the electromagnetic field at the sample;

3. a vector $\vec{t}$ connecting the center of the scattering chamber with the detector.

1.1 Azimuthal rotation of the sample and sample vector

Let $\hat{s}$ be the unit vector of the projection on the X_M, Y_M plane of the vector $\vec{W}$, fixed with the sample and not parallel with the sample normal $\hat{n}_S$, chosen as reference vector for the sample rotations around its normal. Indicating with ϕ_M the angle between $\hat{s}$ and the X_M axis (see Figure 4), we have:

$$\hat{s} = \cos\phi_M \, \hat{\imath}_M + \sin\phi_M \, \hat{j}_M$$

with $\hat{\imath}_M$ and $\hat{j}_M$ the unit vectors of the axes x_M and y_M respectively.

1.2 The vectors of the incident electromagnetic field: $\vec{k}$, $\vec{E}_H$ and $\vec{E}_V$.

Aim of this paragraph is to give the components of the incident electromagnetic field in the manipulator frame of reference, assuming the chamber at an angular position Ψ_C (i.e. the chamber rotated of this angle around X_C) and the manipulator at an angular position θ_M (i.e. the manipulator rotated

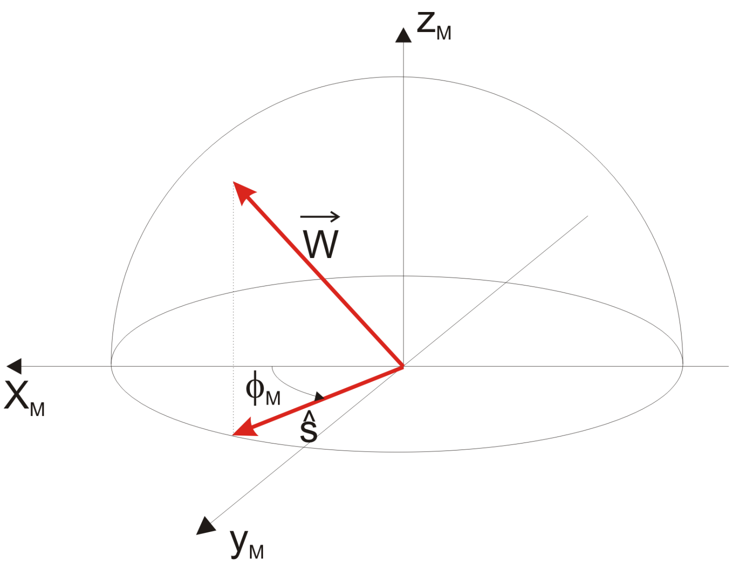

Figure 4: Definition of the azimuthal angle in the manipulator frame of reference.

of this angle around Y_C) together with the definition of the light vector $\vec{k}$ (Figure 5).

The zeros of Ψ_C and θ_M are taken as follows:

- $\Psi_C = 0$ when y_C is parallel to y_L and z_C is parallel to z_L.
- $\theta_M = 0$ when Z_M axis is parallel to Z_C (sample horizontal).

Their sense of rotation is taken according with rotation of a right hand screw advancing along the positive direction of x_C for Ψ_C and ofy_C for θ_M.

The electromagnetic field is general elliptically polarized. In the laboratory reference system the electromagnetic wave propagates along the positive direction of the x_L axis. The elliptical polarity of the light results from the superposition of the two motions along the z_L and y_L axes: be $\vec{E}_H$ and $\vec{E}_V$ the two components of the electric field along the y_L and z_L directions, respectively.

We can write the component of $\vec{E}_H$ and $\vec{E}_V$ in the manipulator reference frame Ω_M:

$$\begin{cases} E_H^x = -\left|\vec{E}_H\right| \sin\Psi_C(-\theta_M) \\ E_H^y = \left|\vec{E}_H\right| \cos\Psi_C \\ E_H^z = -\left|\vec{E}_H\right| \sin\Psi_C \cos(-\theta_M) \end{cases} \quad \begin{cases} E_V^x = \left|\vec{E}_V\right| \cos\Psi_C(-\theta_M) \\ E_V^y = \left|\vec{E}_V\right| \sin\Psi_C \\ E_V^z = \left|\vec{E}_V\right| \cos\Psi_C \cos(-\theta_M) \end{cases} \tag{1}$$

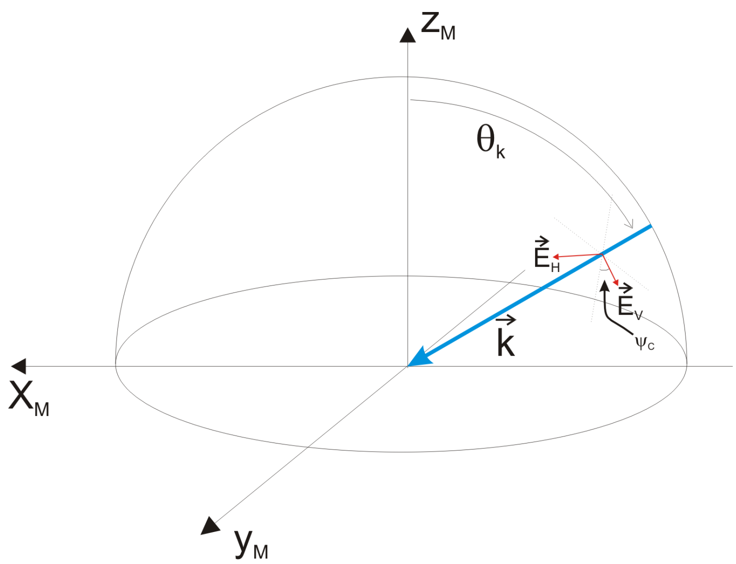

Figure 5: Scheme to calculate the components of the light vector $\vec{k}$ and the electric field components in the manipulator frame of reference.

These equations shows that the chamber rotation Ψ_C changes the position of the polarization ellipse with respect to the sample. In fact for the limit case of horizontal linearly polarized field (left equations) for $\Psi_C = 0$, the electric field is always perpendicular to z_M and parallel to y_M (s polarized radiation), while for $\Psi_C = \pi/2$ the electric field has only x_M and z_M components (p polarized radiation). Both sets of equations must be considered for the elliptically polarized light.

Independently from the chamber rotation Ψ_C, the direction of the light, i.e. the vector $\vec{k}$, is always contained in the x_M-y_M plane. With reference to the Figure 5, θ_k is the angle formed between the direction of incidence and the z axis. θ_k is zero when the light is impinging in the direction of the z_M axis ($\vec{k}$ antiparallel to z_M), and its sense of rotation is positive according with a right hand screw advancing along the positive direction of the y_M axis. The two angles θ_k and θ_M are linked by the relation:

$$\theta_k = \frac{\pi}{2} - \theta_M. \tag{2}$$

Consequently θ_k and ϕ_M, together with Ψ_C, completely define the light incidence conditions (direction and polarization) on the sample.

1.3 Detector position: the $\vec{t}$ vector

The detector features two degrees of angular freedom with respect to the chamber's body, θ_A and ϕ_A, as shown in Figure 2. The detector position must be described in the manipulator frame of reference Ω_M.

The position of a generic point P on a sphere of unit radius is uniquely determined by giving two angles: ψ and θ (see Figure 6). We can define the coordinates in the xyz-reference system by means of the following relations:

$$x = -\sin\psi\ \cos\theta \tag{3a}$$

$$y = \cos\psi\ \cos\theta \tag{3b}$$

$$z = \sin\theta \tag{3c}$$

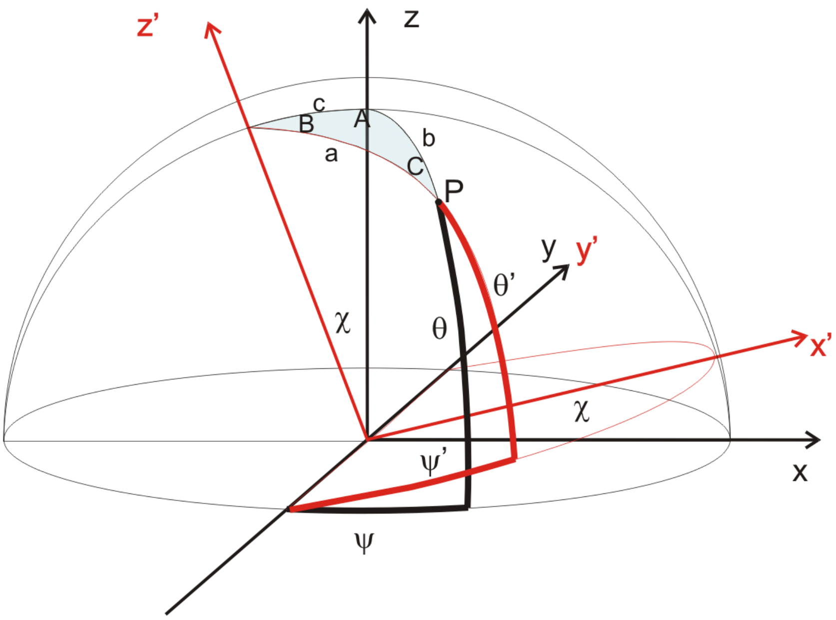

Figure 6: The position of a point on a sphere is defined by two angles. To derive triangular formulae for the spherical triangle ABC, the spherical coordinates ψ, θ, ψ' and θ' of the vertex C are expressed in terms of the sides and angles of the triangle.

In the same way, we can define the angles ψ' and θ', which give the position of P in the $x'y'z'$-frame. We obtain analogous relations:

$$x' = -\sin\psi'\ \cos\theta' \tag{4a}$$

$$y' = \cos\psi'\ \cos\theta' \tag{4b}$$

$$z' = \sin\theta' \tag{4c}$$

The second system of coordinates is obtained from the first one by a rotation around the y axis (see Figure 7):

$$x' = x\cos\chi + z\sin\chi$$
$$y' = y$$
$$z' = z\cos\chi - x\sin\chi$$

or, in matrix form

$$\begin{pmatrix} x' \\ y' \\ z' \end{pmatrix} = \begin{pmatrix} \cos\chi & 0 & \sin\chi \\ 0 & 1 & 0 \\ -\sin\chi & 0 & \cos\chi \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix} \tag{5}$$

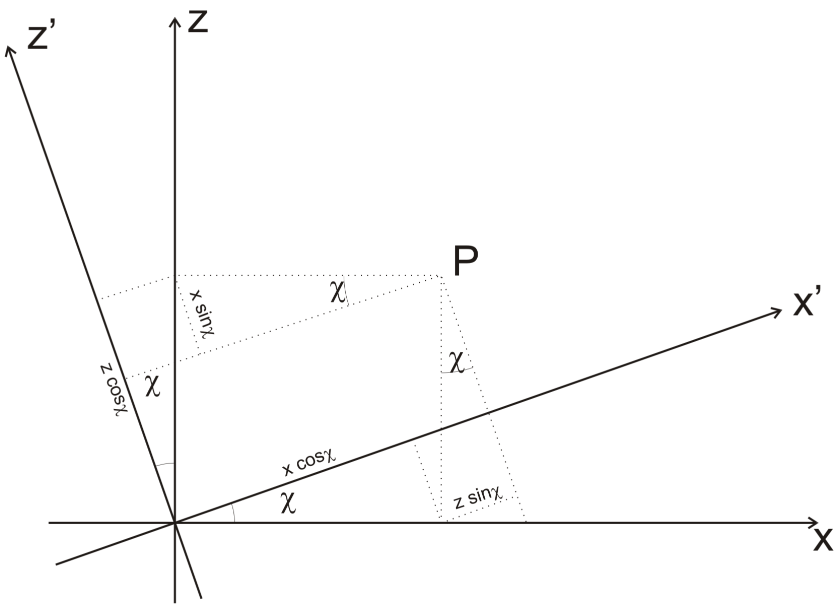

Figure 7: Coordinates of point P in the reference frame xyz e $x'y'z'$

Combining the equations (4) and (5), we obtain the relations between the two reference systems:

$$-\sin\psi' \ \cos\theta' = -\sin\psi \ \cos\theta \ \cos\chi + \sin\theta \ \sin\chi \tag{6a}$$
$$\cos\psi' \ \cos\theta' = \cos\psi \ \cos\theta \tag{6b}$$
$$\sin\theta' = \sin\psi \ \cos\theta \ \sin\chi + \cos\chi \ \sin\theta. \tag{6c}$$

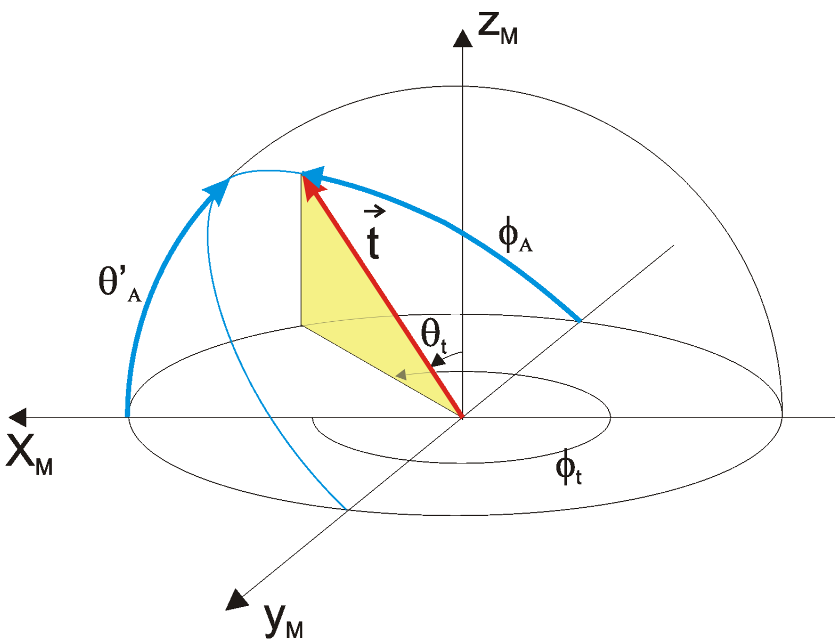

Figure 8: The unit vector of the detector in the Ω_M frame of reference and the relation between the components in Ω_M the angle θ'_A and ϕ_A of the detector axes.

The particular case of the experimental chamber at BEAR is depicted schematically in Figure 8. The position of the detector is described by the vector $\vec{t}$. In the manipulator frame of reference the analyzer goes around circular trajectories described by the blue line, where θ'_A is the angle between the x_M-y_M plane and the plane of the trajectory. The value of $\theta' A$ is given by the combination of the setting of the manipulator angle θ_M and the detector angle θ_A:

$$\theta'_A = \theta_A - \theta_M \tag{7}$$

Applying the angle definition expressed in Figure 6, compared with Figure 8, in the case of the experimental chamber at BEAR we have:

$$\begin{aligned}
&\psi = \phi_t + \frac{\pi}{2} \\
&\psi' = \phi_A \\
&\theta = \frac{\pi}{2} - \theta_t \\
&\theta' = 0 \\
&\chi = \theta'_A
\end{aligned} \tag{8}$$

which, applied into equations (6), give:

$$-\sin\phi_A = -\cos\theta'_A\ \sin\left(\phi_t+\frac{\pi}{2}\right)\ \cos\left(\frac{\pi}{2}-\theta_t\right)+\sin\theta'_A\ \sin\left(\frac{\pi}{2}\right) \tag{9a}$$

$$\cos\phi_A = \cos\left(\phi_t+\frac{\pi}{2}\right)\ \cos\left(\frac{\pi}{2}-\theta_t\right) \tag{9b}$$

$$0 = \sin\theta'_A\ \sin\left(\phi_t+\frac{\pi}{2}\right)\ \cos\left(\frac{\pi}{2}-\theta_t\right)+\cos\theta'_A\ \sin\left(\frac{\pi}{2}-\theta_t\right) \tag{9c}$$

$$\begin{aligned}-\sin\phi_A &= -\cos\phi_t\sin\theta_t\cos\theta'_A+\cos\theta_t\sin\theta'_A\\ \cos\phi_A &= \sin\phi_t\ \sin\theta_t\\ 0 &= \sin\theta_t\ \sin\theta'_A\ \cos\phi_t+\cos\theta_t\ \cos\theta'_A\end{aligned}$$

Solving for ϕ_A and θ'_A, we obtain:

$$\boxed{\begin{aligned}\theta'_A &= -\arctan\left(\frac{\cot\theta_t}{\cos\phi_t}\right)\\ \phi_A &= \arccos\left(\sin\phi_t\ \sin\theta_t\right)\end{aligned}} \tag{10}$$

The inverse transformations are obtained applying the inverse of matrix (5):

$$\begin{pmatrix}\cos\chi & 0 & \sin\chi\\ 0 & 1 & 0\\ -\sin\chi & 0 & \cos\chi\end{pmatrix}^{-1} = \begin{pmatrix}\cos\chi & 0 & -\sin\chi\\ 0 & 1 & 0\\ \sin\chi & 0 & \cos\chi\end{pmatrix} \tag{11}$$

We find

$$-\sin\left(\phi_t+\tfrac{\pi}{2}\right)\ \cos\left(\tfrac{\pi}{2}-\theta_t\right) = -\sin\phi_A\ \cos\theta'_A \tag{12}$$

$$\cos\left(\tfrac{\pi}{2}-\theta_t\right)\ \cos\left(\phi_t+\tfrac{\pi}{2}\right) = \cos\phi_A \tag{13}$$

$$\sin\left(\tfrac{\pi}{2}-\theta_t\right) = -\sin\phi_A\ \sin\theta'_A \tag{14}$$

that yields to

$$\boxed{\begin{aligned}\theta_t &= \arccos\left(-\sin\phi_A\ \sin\theta'_A\right)\\ \phi_t &= \arctan\left(\cot\phi_A\ \cos\theta'_A\right)\end{aligned}} \tag{15}$$

We must fix the intervals for θ_t and ϕ_t. The movement of ϕ_t is limited by the presence of the manipulator. The limits for ϕ_t are:

$$\begin{aligned}0 \le \phi_t &\le \frac{\pi}{4}\\ \frac{3\pi}{4} \le \phi_t &\le 2\pi\end{aligned}$$

and for θ_t is

$$0 \leq \theta_t \leq \frac{\pi}{2}$$

Each movement must be expressed in terms of the angles θ_t and ϕ_t in the system of reference of the sample.