New Experimental Methods for Perturbation Crystallography.
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Chapter 2

Theory

2.1 Introduction

Since their discovery more than 100 years ago by the Curie brothers\cite{1,2} (Pierre and Jacques), piezoelectric materials have been studied at the macroscopic scale and several theories have been developed to explain piezoelectricity. The existence of any theory at microscopic, i.e. atomic scale, is rather limited\cite{2} whereas the prediction of the magnitude of piezoelectricity based on ab-initio principles is becoming available\cite{3,4}. However, recent studies have investigated the effect at the microscopic scale\cite{5,6} and are mainly focused on the well-known and commercially used crystals of LiNbO$_3$, KTiOPO$_4$, AgGaS$_2$ and quartz.

This chapter will discuss briefly piezoelectricity (§2.2) and its properties in relation to X-ray diffraction (§2.3), followed by a description of the theory of X-ray sources (§2.4) with an emphasis on the synchrotron X-ray source of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France.

2.2 Piezoelectricity

The piezoelectric effect, as will be explained in the following section, can be divided into two distinct effects: the direct piezoelectric effect and the converse piezoelectric effect.

2.2.1 Direct piezoelectric effect

The phenomenon that certain crystals experience a change of the electric polarisation and develop electric charges on opposite crystal faces upon application of a mechanical stress is known as the direct piezoelectric effect.
In general all non-centrosymmetric crystals, with the exception of the cubic class 432, are piezoelectric. Hence, twenty point groups show piezoelectric behaviour. However, the absence of a centre of symmetry is an essential but not a sufficient requirement, because the magnitude and direction of the piezoelectric effect depend also on the direction of the applied stress\[^{12}\] and the contents of the material\[^{13}\] as is shown in Figure 2-1 and Figure 2-2, respectively. An unstressed ferroelectric crystal (Fig. 2-2a) with a spontaneous polarisation is stressed (Fig. 2-2b), resulting in an induced polarisation $\Delta P$ with a magnitude proportional to the applied stress. An unstressed non-ferroelectric crystal with a three-fold symmetry is shown in Figure 2-2c. Here the arrows represent dipole moments, where each set of three arrows represents a planar group of ions denoted by $(A^+, B^3)$, with a $B^3$ ion at each vertex. The sum of the three dipole moments at each vertex is zero and no spontaneous polarisation occurs. However, when the crystal is stressed the three-fold symmetry will be broken and polarisation occurs in the indicated direction (Fig. 2-2d).

The magnitude of the induced electric polarisation is proportional to the applied stress $\sigma$, and is given in a first approximation, under isothermal and isobaric conditions, by

$$ P = d\sigma, \quad (2-1) $$

where $d$ is the piezoelectric tensor\[^{14}\].

Using the Einstein summation convention, Equation 2-1 can be written as

$$ P_i = d_{i\mu}\sigma_\mu, \quad (i, j, k = 1, 2, 3), \quad (2-2) $$

where $d_{i\mu}$ are the piezoelectric moduli. This means that when a general stress $\sigma_\mu$ acts on a piezoelectric crystal each component of the polarisation $P_i$ is linearly related to all the components of $\sigma_\mu$.

When a letter suffix occurs twice in the same term, summation from 1 to 3 with respect to that suffix is understood automatically. For example:

$$ p_1 = \sum_{j=1}^{3} T_{1j} q_j, \quad p_2 = \sum_{j=1}^{3} T_{2j} q_j, \quad p_3 = \sum_{j=1}^{3} T_{3j} q_j, \quad p_i = \sum_{j=1}^{3} T_{ij} q_j \quad (i=1,2,3) $$

$$ p_i = T_{ij} q_j, \quad (i,j=1,2,3) $$
Figure 2-1: Different application of stress upon a piezoelectric crystal with their respective axis of induced polarisation.

Figure 2-2: Piezoelectric effect versus contents of material: a: Unstressed ferroelectric crystal; b: Piezoelectric effect caused by applying a stress to the unstressed ferroelectric crystal which produces a change in the polarisation by $\Delta P$, the induced piezoelectric polarisation; c: A non-ferroelectric crystal with a zero net dipole moment for the threefold symmetry axis; d: When a stress is applied the threefold symmetry breaks and a non-zero net dipole moment occurs (from Kittel).
Furthermore, it should be noted that as the state of stress is identified by a second-rank tensor with nine components and the polarisation of a crystal, being a vector, is identified by three components, $d_{jk}$ is a third-rank tensor with 27 components.

Since $d_{jk}$ is symmetric in $j$ and $k$ a reduction of the components can be obtained. Elimination of one of each set $jk$ in the symmetric $d_{jk}$ results in 18 elements.

\[
\begin{array}{ccccccc}
    d_{11} & d_{12} & d_{13} & d_{21} & d_{22} & d_{23} & d_{31} & d_{32} & d_{33} \\
    d_{21} & d_{22} & d_{23} & d_{31} & d_{32} & d_{33} & d_{12} & d_{13} & d_{23} \\
    d_{31} & d_{32} & d_{33} & d_{13} & d_{23} & d_{33} & d_{12} & d_{13} & d_{23} \\
\end{array}
\]

With this new set of $d_{jk}$ a further simplification can be obtained by changing from tensor notation to matrix notation, giving a clearer and more convenient mathematical approach when calculating particular problems. Using Voigt's convention the matrix elements are set as follows:

<table>
<thead>
<tr>
<th>Tensor notation $(j, k)$</th>
<th>11</th>
<th>22</th>
<th>33</th>
<th>21</th>
<th>32</th>
<th>31</th>
<th>12</th>
<th>21</th>
</tr>
</thead>
<tbody>
<tr>
<td>Matrix notation $(j)$</td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>4</td>
<td>5</td>
<td>6</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Thus, for example $d_{22}=d_{211}$ and $d_{12}=2d_{122}$. For consistency, the suffix notation of the stress components in Equation 2-2 will change to the matrix notation as follows

\[
\begin{bmatrix}
    \sigma_{11} & \sigma_{12} & \sigma_{13} \\
    \sigma_{21} & \sigma_{22} & \sigma_{23} \\
    \sigma_{31} & \sigma_{32} & \sigma_{33}
\end{bmatrix}
\Rightarrow
\begin{bmatrix}
    \sigma_1 & \sigma_2 & \sigma_3 \\
    \sigma_4 & \sigma_5 & \sigma_6
\end{bmatrix}.
\]  

(2-3)

Rewriting Equation 2-2 into the new notation gives the matrix notation

\[P_i = d_{ij} \sigma_j \quad (i = 1, 2, 3; j = 1, 2, \ldots 6),\]  

(2-4)

where the piezoelectric elements $d_{ij}$ are given by

\[
\begin{pmatrix}
    d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\
    d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\
    d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36}
\end{pmatrix}
\]

(in CN).  

(2-5)

The symmetry in the $jk$ elements of $d_{jk}$ is the consequence of the symmetrical tensor, when any second-rank tensor is expressed as the sum of a symmetrical and an anti-symmetrical tensor. The introduction of a factor of 2 allows Equation 2-4 to be written in a compact way without any factor.
In Appendix A an overview is given of the non-zero piezoelectric moduli in the acentric point groups.

2.2.2 Converse piezoelectric effect

When an external electric field is applied upon a piezoelectric crystal a strain within the crystal appears. This is the so-called converse piezoelectric effect.

From thermodynamics it follows that the coefficients for the converse piezoelectric effect are numerically equal to the coefficients for the direct effect. Therefore, the mathematical relation between the applied external electric field $E_i$, and the strain $\varepsilon_{jk}$, of the converse piezoelectric effect is given by

$$\varepsilon_{jk} = d_{jk} E_i \quad (i, j, k=1, 2, 3). \quad (2-6)$$

Using the $jk$ symmetry of $\varepsilon$ and $d$ and the defined matrix notation as given in §2.2.1, Equation 2-6 can be written (using Voigt’s notation) as

$$\varepsilon_j = d_{ij} E_i \quad (i=1, 2, 3; j=1, 2, \ldots 6), \quad (2-7)$$

where $d_{ij}$ is as defined in Equation 2-5.

---

* An elaborate explanation on the equality of the coefficients of the converse and direct piezoelectric effect can be found in Nye[4]. The figure shows the relations between the thermal, electrical and mechanical properties of a crystal. The names of the properties and the variables are given. The tensor rank of the variables is shown in round brackets and the tensor rank of the properties in square brackets (from Nye).
2.2.3 Crystal symmetry

A further reduction of the piezoelectric tensor is possible due to the crystal symmetry, if present. Some of the piezoelectric moduli might be zero, equal to or linearly related to a symmetry-related modulus. Therefore the final $d_{ij}$ tensor will contain less than 18 independent moduli.

2.3 X-ray Diffraction

The studies on piezoelectricity carried out at the macroscopic level during the past decades focused basically on the piezoelectric constants. In most of these cases the direct piezoelectric effect was used to measure these constants macroscopically.

However, over the last 25 years, piezoelectricity is being investigated increasingly by means of X-ray measurements which allow the study of these effects at the microscopic level, where little is known about the piezoelectric effect.

X-ray diffraction is an excellent technique to study piezoelectricity at the atomic scale, because it allows the study of the three distinct effects which can be observed when a piezoelectric crystal is subjected to an external electric field. These effects are:

1. A change in the Bragg angle, which can be used to determine a piezoelectric constant of the piezoelectric tensor.
2. A change in integrated intensity associated to possible changes of the electron-density distribution or atomic-positional parameters and
3. A change in rocking curve width, which relates to changes in the mosaic spread.

It must be stressed that these three effects are very small in magnitude and measuring the effects with good counting statistics is very time consuming. These experimental difficulties are, or rather were, until recently the main reason for the sporadic publications. Recent developments in measuring these small effects will be discussed in more detail in the following chapters.

2.3.1 Change in Bragg angle

The possibility of measuring piezoelectric constants by means of X-ray diffraction was first shown by Bhalla et al.\textsuperscript{[15]}, whereas Barsch\textsuperscript{[16]} presented the first theoretical overview for the determination of the piezoelectric constants from X-ray diffraction data.

For a non-perturbed piezoelectric crystal, Bragg's form of the condition for constructive reflection of an incident X-ray beam applies and for a set of lattice planes with Miller indices $hkl$ it is given by

$$\sin \theta = \frac{n\lambda}{2d_{hkl}}, \quad (2-8)$$
where $d_{hkl}$ is the interplanar spacing. Applying an electric field to a piezoelectric crystal induces an elastic strain (converse piezoelectric effect, Eq. 2-7). This means that the interplanar $d_{hkl}$ for a certain set of Miller planes $(hkl)$ changes into $d'_{hkl}$. Keeping the incoming X-ray beam at the same wavelength a change of the Bragg angle $\theta$ by an amount $\Delta\theta$ will occur. Furthermore, Graafsm\textsuperscript{11,18} observed an additional effect on the change of the Bragg angle which is caused indirectly by the piezoelectric effect. This additional effect stems from a rotation of the crystal due to the constraint of the crystal mount and the applied electric field. Therefore, the observed $\Delta\theta_{\text{obs}}$ as a response to the electric field consists of two contributions

$$\Delta\theta_{\text{obs}} = \Delta\theta_\theta + \Delta\theta_{\text{rot}},$$

(2-9)

where a change of the unit cell causes a change of the Bragg angle $\Delta\theta_\theta$ and the rotation of the entire lattice is $\Delta\theta_{\text{rot}}$.

**Piezoelectric contribution**

Barsch\textsuperscript{16} describes how the Bragg angle $\theta_\theta$ for a certain reflection changes by an amount $\Delta\theta_\theta$ as is given by

$$\Delta\theta_\theta = -\tan\theta_\theta \sum_{i=1}^{3} \sum_{j=1}^{3} h_{i,j} e_{i,j}$$

$$= -E \tan\theta_\theta \sum_{k=1}^{4} \sum_{l=1}^{4} e_{k,l} h_{k,l} d_{k,l},$$

(2-10)

where $E$ is the magnitude of the electric field, $e_{i,k}$ and $h_{i,j}$ are the directional cosines of the electric field and the diffraction vector $h_{i,j}$, respectively. As can be seen, the shift in the Bragg angle is a function of $\tan\theta$, implying that high-order reflections will show larger shifts than low-order reflections.

**Rigid rotation**

In contrast to the piezoelectric contribution, the rotational contribution is non-material specific and is independent of $\theta$. It depends purely on the strength of the applied electric field and the way the sample is mounted. As the strain in the crystal is proportional to the applied electric field, a rigid mounting of the sample does not allow any shape deformations and the strained crystal responds by twisting, giving an extra change, $\Delta\theta_{\text{rot}}$, to the observed angle of diffraction. Hence, mounting crystals in such a way that shape deformations are allowed will decrease the rigid rotation significantly.
2.3.2 Change in integrated intensities

When X-ray radiation interacts with a crystal, scattering or diffraction of the X-rays occurs. The resulting diffraction pattern is unique for each material and represents the internal atomic occupation and structure.

The type and position of the atoms in the crystal's unit cell define the structure factor for a particular reflection.

\[ F(hkl) = \sum_{i=1}^{n} g_i e^{i\mathbf{h} \cdot \mathbf{r}_i} \]

\[ = \sum_{i=1}^{n} f_i \cdot T_{ij} \cdot e^{2\pi i h \cdot r_{ij}}, \quad (2-11) \]

where \( f_i \) is the atomic form factor, \( T_{ij} \) the atomic displacement parameter function, \( hkl \) represents the Miller indices of a reflection and \( \mathbf{r}_{ij} \) are the fractional co-ordinates of an atom. Equation 2-11 can also be expressed using the electron-density distribution function \( \rho_i \).

\[ F_h = \int \rho_i e^{2\pi i h \cdot \mathbf{r}} d\mathbf{r}, \quad (2-12) \]

where \( \mathbf{h} \) denotes the scattering vector and \( \mathbf{r} \) is the positional vector.

The observed intensity for a certain reflection is related to the magnitude of the structure factor by

\[ I_{\omega h} = k|F_h|^{2}. \quad (2-13) \]

where \( k \) is a scale factor and \( F^* \) is the complex conjugate of \( F \). Throughout this work the intensity expression in Equation 2-13 will be used. For reasons which will be explained in Chapter 6, Equation 2-13 is a simplified form and, in fact, the experimentally observed intensity depends also on other factors such as the Lorentz factor, polarisation, absorption and extinction effects.

Application of an electric field to a piezoelectric crystal induces a change in the integrated intensity of Equation 2-13 by a factor of

\[ \Delta I_h = \Delta (F_h F_h^*). \quad (2-14) \]

From the changes in integrated intensities, shifts in the atomic positions or changes in \( g_s \) (Eq. 2-11) can be calculated using an appropriate structure factor calculation program.

Internal and external strain

The strain in a piezoelectric crystal caused by the application of an electric field can be divided into two strain effects which both influence the structure factor in a different manner.
The first effect is the so-called elastic or external strain which describes the (elastic) deformation of the crystal. This becomes visible in an X-ray diffraction experiment as a change in Bragg angle (§2.3.1). Since the magnitude of the structure factor depends on the Bragg angle via the atomic form factor and the atomic displacement parameters, small changes in the structure factor are to be expected. Furthermore, as the external strain for atomic structures does not affect the fractional co-ordinates of the atoms in the unit cell, the exponential form in the structure factor will not be affected. However, this does not hold for structures containing or consisting of rigid bodies. Although the fractional co-ordinates of the centre of mass for a rigid body will not change when the crystal is strained, the fractional co-ordinates of the rigid body’s atoms do change. Hence, the change in the structure factor, which is the combined effect of the change in shape and fractional co-ordinates, will be significantly larger in comparison to that of a non-rigid body structure.

The second strain effect involves the change of the atomic positions within the unit cell and is referred to as the internal strain. However, contrary to the external strain, the internal strain will affect the exponent in the structure factor since the atomic fractional co-ordinates change. Therefore, the change of the structure factor’s magnitude will be significantly larger than the one induced by the external strain effect.

When the external strain is assumed not to affect the structure factor significantly, an experimental separation of both effects can be obtained. Measuring the changes in the Bragg angle give information on the external strain whereas differences in intensities give information on the internal strain.

2.3.3 Change in rocking curve width

Topography studies showed that the application of an external electric field upon a piezoelectric crystal might change the crystal perfection, especially the mosaicity. This can be observed in a diffraction experiment as a change in the rocking curve width. However, this phenomenon is not the subject of this work, since the mosaicity of a crystal is at the meso-macroscopic scale rather than at the microscopic, i.e. atomic, scale.

2.4 X-ray Sources

A wide variety of X-ray sources can be found nowadays ranging from small laboratory equipment to large X-ray facilities. Their use depends on the different needs of the experimentalist such as ability to tune energy, high brilliance, time structure and polarisation of the X-ray beam. Furthermore, X-ray sources can be found not only in scientific institutions, but they are also common in use in the fields of medicine (e.g. X-ray photos/imaging), astrophysics (e.g. X-ray telescopes), and industry (e.g. thickness measurements of metals and X-ray machines at airports). However, the latter applications of X-ray sources will not be discussed, though the production of X-rays (as explained in the following sections) is the same.
2.4.1 Conventional X-ray sources

Conventional X-ray sources such as the X-ray tube and the rotating anode, are common and widely used in laboratories as routine data-acquisition instrument sources, although the low brilliance and the discrete wavelength limits their flexibility.

The principle for creating an X-ray beam is fairly simple. A cathode is heated in such a way that electrons are emitted and travel in a vacuum to a steady or rotating anode due to the high potential difference (≈2-6 kV). The anode is usually made of Cu or Mo depending on the requirement of the flux and wavelength. A highly energetic electron may remove a K-shell electron of the anode-atom thereby creating a hole in the K-shell. Electrons from higher energy shells refill more or less instantaneously the K-shell with the emittance of radiation corresponding to the excess in energy. The emitted radiation has a wavelength characteristic for the anode material. The main spectral lines of an X-ray tube, the $K_{\alpha}$ and $K_{\beta}$ lines with an intensity distribution of 6 to 1, are due to electron transitions from L and M to the K-shell, respectively. Furthermore, each K line consists actually of at least two lines with a small difference in the wavelength, due to the slightly different energy levels within the L and higher electron shells. The intensity distribution is about 2 to 1 for $K_{\alpha 1}$ and $K_{\alpha 2}$, for example. Table 2-1 gives the possible energies, i.e. wavelengths for a Cu and Mo anode. Note that other emission lines are possible by using anode materials such as Ni, Ag, and Fe, although they are not in common use. On the other hand W is widely used in high-energy X-ray machines which are used for medical examinations and industrial applications.

<table>
<thead>
<tr>
<th>Anode</th>
<th>Spectral line</th>
<th>Wavelength [Å]</th>
<th>Energy [keV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>$K_{\beta}$</td>
<td>1.3922</td>
<td>8.905</td>
</tr>
<tr>
<td>Cu</td>
<td>$K_{\alpha 1}$</td>
<td>1.5406</td>
<td>8.0478</td>
</tr>
<tr>
<td>Cu</td>
<td>$K_{\alpha 2}$</td>
<td>1.5444</td>
<td>8.0278</td>
</tr>
<tr>
<td>Mo</td>
<td>$K_{\beta}$</td>
<td>0.6323</td>
<td>19.608</td>
</tr>
<tr>
<td>Mo</td>
<td>$K_{\alpha 1}$</td>
<td>0.7093</td>
<td>17.4793</td>
</tr>
<tr>
<td>Mo</td>
<td>$K_{\alpha 2}$</td>
<td>0.7136</td>
<td>17.3743</td>
</tr>
</tbody>
</table>

In most cases the $K_{\alpha 1}$ and/or $K_{\alpha 2}$ lines are selected, by using a monochromator, due to their steep spectrum profile and high intensity. In comparison, the $K_{\beta}$ is low in intensity. The X-ray spectrum contains white radiation, also known as bremsstrahlung, which stems from multiple electron collisions within the anode metal as not all of the accelerated electrons are stopped fully in a single collision. Consequently, a continuous spectrum with relatively high intensities at small wavelengths in comparison to large wavelengths will be formed. White radiation is not anode-material dependant and depends on the machine voltage. In many cases the $K_{\beta}$ line and white radiation are absorbed by a Be-window, which separates the vacuum and the laboratory environment, the crystal monochromator or (metal) filters.
When high brilliance and the ability to tune the wavelength is of importance synchrotron radiation is an appropriate choice.

### 2.4.2 Synchrotron X-ray sources

A synchrotron is a state-of-the-art machine for the production of a polarised X-ray beam with high brilliance and a wide energy range spectrum. Furthermore, due to the principles of a synchrotron source, a pulsed-time structure is present in the X-ray beam, which allows time-resolved studies down to the picosecond regime\(^\text{[29]}\).

All synchrotron sources are based on the same principle: When a charged particle changes momentum, electromagnetic (EM) radiation is emitted. The emission of EM radiation for electrons with \(v/c<<1\) (non-relativistic electrons) is isotropically distributed around the radiating electrons, where \(v\) is the velocity of the electron and \(c\) the speed of light, whereas the EM emission stemming from relativistic electrons (\(v/c=1\)) is sharply peaked in the direction of motion of the radiating electrons.

Since the invention of a synchrotron source\(^\text{[29]}\) in 1946, a continuous improvement in brilliance and energy spectrum has been obtained for different generations of synchrotron sources. An overview of the increase in brilliance during the last decades for several X-ray sources is shown in Figure 2-3.

All the experimental work presented in this work has been performed at the European Synchrotron Radiation Facility (ESRF), a third generation synchrotron source, situated in Grenoble, France. The discussion of the principles of synchrotron sources will be mainly focused on the ESRF, although the basics of synchrotron radiation are applicable to all synchrotron sources.

Synchrotron sources are divided into three distinct groups or generations. The first two generations of synchrotron sources are rather limited in brilliance (e.g. Hasylab, NSLS, SSRL and SRS) and the divergence of the X-ray beam is large. The first generation synchrotrons are merely parasitic to high-energy sources, whereas the second-generation synchrotrons are dedicated sources. Despite the limitation in the delivered brilliance, these machines are preferred in comparison to the conventional sources for some applications. However, by the implementation of improved insertion devices in third generation sources, as will be explained in the next sections, the brilliance was increased by a factor of 2 to 3 in magnitude and a smaller divergence was obtained (ESRF, APS and SPRing-8).

Synchrotrons use electrons (in some cases positrons) to generate an X-ray beam. The electrons are thermally emitted by a klystron, in much the same way as a cathode in a conventional source, and accelerated in a linear accelerator (LINAC) to a given energy. The accelerated electrons are then transferred to a ring or oval-type accelerator, called a booster. The booster sweeps up the electron’s energy, by means of radio frequency (RF) cavities, to the machine’s working energy. Finally the electrons are transferred to the storage ring. Here the electrons run in circles (closed orbit) confined by focusing and bending magnets. As a result of this circular confinement the momentum of the
electrons is continuously changed and production of EM radiation, including X-rays, occurs. Furthermore, insertion devices can be installed in the storage ring allowing the production of X-rays with certain properties such as specific distribution of X-ray energies and polarisation direction. The radiation, as is directed by the forward X-ray emission of the relativistic electrons, enters the experimental floor in specially designed scientific stations where experiments can be performed.

**Figure 2-3:** History of brilliance versus X-ray sources (courtesy ESRF).
An overview of the general properties of the storage ring will be discussed, as a reference for further use in the next paragraphs.

The time of circulation, the orbit time $T$, for relativistic electrons is given by

$$ T = \frac{L}{c}, $$

where $L$ is the circumference of the storage ring, which depends on the machine's energy and the magnetic field strength of the bending magnets (see §Bending magnet). Since the electrons lose some of their energy when they cycle in the storage ring due to emission of radiation and/or collision with ions (since an ideal vacuum is unattainable), energy is replenished by a RF field. As a continuous electron stream in the storage ring cannot be accelerated by RF cavities, bunches of electrons are used. The maximum number of bunches is defined by

$$ N = \frac{v_n L}{c}, $$

where $v_n$ is the RF frequency.

Furthermore, the relativistic electrons can be described as the ratio of the electron energy $E$ and their rest energy $m_0c^2$ by

$$ \gamma = \left[1 - \left(\frac{v}{c}\right)^2\right]^{-1} = \frac{E}{m_0c^2}, $$

or

$$ \gamma = 1957E \quad (E \text{ in GeV}), $$

where $m_0$ is the rest mass of an electron.

2.4.3 ESRF

The ESRF was the first third generation synchrotron built and operates at an energy of 6 GeV. Figure 2-4 gives a general overview of the synchrotron source of the ESRF. The electrons emitted by the klystron are accelerated in the LINAC (16 m length) to an energy of 200 MeV. After transfer of these high-energy electrons into the booster, which is a 10 Hz cycling synchrotron with circumference of 300 m containing alternately focusing and bending magnets with RF cavities, an acceleration up to 6 GeV is induced. Finally, the electrons are transferred to the storage ring where the electrons will cycle for several hours at an energy of 6 GeV. The ring has a circumference of 844 m and the beam cycles thus every 2.81 µsec (Eq. 2-15). There are 64 beam ports where an X-ray beam can be taken from the various X-ray sources (bending magnets or insertion devices). The machine can be run in different modes which defines the X-ray beam structure. These modes
Chapte rr  2
consis tt  o f  differen t packin g  o f  th e  electron s  i n  th e  bunche s  o f  th e  storag e  rin g  an d  set s  th e  bea m
current ,, bea m  deca y  an d  pulsed-tim e  structur e  parameters . Th e  storag e  rin g  i s  divide d  int o  99 2
bunche ss  wher e  theoreticall y  eac h  bunc h  ca  n  contai n  a  certai n  amoun t o f  electrons . Tabl e  2- 2  show s
the differen t mode s  use d  a t th e  ESRF. Dependin g  o n  th e  selecte d  machin e  mod e  som e  o f  these
bunche ss  ar e  fille d  wit h  electrons , whil e  other s  ar e  kep t empty . I t shoul d  b e  note d  tha t th e  modes  an
d thi eir  propertie s  use d  a t othe r  synchrotron s  ar e  differen t fro m  thos e  use d  a t th e  ESRF.

At the ESRF bending magnets and two different types of insertion devices, either wigglers or
undulators, are situated in the storage ring and produce their characteristic X-ray beams.

**Bending magnet**

A bending-magnet is primarily used for bending the electron-beam path in the storage ring in order
to have a more or less circular path (closed orbit) and is situated at the curved sections of the
storage ring.

<table>
<thead>
<tr>
<th>Mode</th>
<th>Current [mA]</th>
<th>Half life-time [hours]</th>
<th>Filling</th>
</tr>
</thead>
<tbody>
<tr>
<td>2/3 fill</td>
<td>200</td>
<td>55</td>
<td>2/3 filled, 1/3 empty</td>
</tr>
<tr>
<td>2x1/3 fill</td>
<td>200</td>
<td>55</td>
<td>2x1/3 filled separated by 1/6</td>
</tr>
<tr>
<td>Hybrid mode 1</td>
<td>200</td>
<td>35</td>
<td>2/3 filled, 1 bunch</td>
</tr>
<tr>
<td>Hybrid mode 2</td>
<td>200</td>
<td>30</td>
<td>2/3 filled, 2 bunches opposite and equally separated</td>
</tr>
<tr>
<td>Hybrid mode 4</td>
<td>200</td>
<td>30</td>
<td>2/3 filled, 4 bunches opposite and equally separated</td>
</tr>
<tr>
<td>16 bunch</td>
<td>90</td>
<td>10</td>
<td>16 bunches at equal distance</td>
</tr>
<tr>
<td>Single bunch</td>
<td>16</td>
<td>6</td>
<td>1 bunch</td>
</tr>
</tbody>
</table>

As has been mentioned before, the change in moment of an electron will generate X-rays.
Therefore, bending magnets are also used as X-ray sources, although they give a lower brilliance
than the insertion devices as will be discussed in the next paragraphs. A schematic plot of a
bending-magnet is shown in Figure 2-5. A bunch of electrons enters the magnetic field of the
bending-magnet and the path of the bunch will be curved due to the Lorentz force $F$ on the electrons
in the bunch. Hence, the radius $\rho$ is defined as

$$\rho = \frac{m_e v \gamma}{F} = \frac{m_e c \gamma}{eB},$$

or
Figure 2-4: Overview of the ESRF; Linac or preinjector (1), Booster (2), Transfer line (3) (courtesy ESRF).

\[ \rho = \frac{3.336E}{B} \quad (\rho \text{ in m, } E \text{ in GeV and } B \text{ in T}). \]  (2-19)
The wavelength corresponding to the critical energy $\varepsilon_c$, defined as the mean energy such that half of the radiated power is at energies larger and half at energies smaller than $\varepsilon_c$, is related to $\rho$ and $\gamma$ by

$$\lambda_c = \frac{4\pi \rho}{3\gamma^3}$$

or by substituting Equations 2-18 and 2-19

$$\lambda_c = \frac{18.64}{BE^2} \quad (\lambda_c \text{ in } \AA, B \text{ in } T \text{ and } E \text{ in } \text{GeV}). \quad (2-20)$$

The vertical emission angle or opening angle $\psi$ of the photon beam is defined as

$$\psi = \frac{1}{\gamma}. \quad (2-21)$$

The intensity of the photon beam as a function of wavelength integrated over the vertical emission angle can be expressed by

$$H(\lambda) = 1.256 \cdot 10^{10} kG(y)\gamma \quad (H \text{ in photons } s^{-1} \text{ mrad}^{-1} \text{ mA}^{-1}) \quad (2-22)$$

with

$$G(y) = \int_y^\infty K_n(t)dt \quad (y = \lambda_y/\lambda_c \text{, and the bandwidth } k \text{ equal to } \Delta \lambda/\lambda_c),$$

where $G(y)$ and the modified Bessel function of the second kind $K_{3/3}$ have been tabulated by Winick. For a 0.1% bandwidth this gives

$$H(\lambda) = 1.256 \cdot 10^{7} G(y)\gamma \quad (H \text{ in photons } s^{-1} \text{ mrad}^{-1} \text{ mA}^{-1}) \quad (2-23)$$

The horizontal cross-section of the X-ray beam will be large due to the bending radius of the electron path.

*Figure 2-5: Principle of a bending-magnet (courtesy ESRF).*
Wiggler

A wiggler is an insertion device which can be implemented in a straight section of the storage ring between two BM's. It consists of series of magnets arranged in such a way that when a bunch of electrons in the storage ring passes through, its path will be sinusoidal, with a period and amplitude according to the magnetic parameter of the wiggler (Fig. 2-6).

![Diagram of electron bunch and X-ray emission](image)

**Figure 2-6: Principle of a wiggler and undulator.** For both insertion devices different arrangements of the magnetic field exist which influences the emission cones, and hence the interference of the X-rays (courtesy ESRF).

The optical properties of the wiggler is given by the parameter $K$ as

$$K = \frac{e}{2\pi m c} \lambda_{osc} B \quad (\lambda_{osc} \text{ in cm and } B \text{ in T})$$

or

$$K = 0.934 \lambda_{osc} B, \quad (2-24)$$

where $\lambda_{osc}$ is the wiggler’s magnetic period. It should be noted that for a wiggler $K >> 1$, $\lambda_{osc} = 7.0$ cm and $B = 0.85$ T are typical values used.

The wavelength depends on $K$ and is given by

$$\lambda_{\gamma}(\theta) = \left( \frac{\lambda_{c, \theta=0}}{1 - \frac{\gamma^2 \theta^2}{K^2}} \right), \quad (2-25)$$

where $\theta$ is the angle of observation with respect to the radiation off-axis and $\lambda_{c, \theta=0}$ is the critical wavelength of Equation 2-20. Hard X-rays are radiated along the axis and softer ones at an angle $\theta$. The radiation fan of the wiggler contains a continuous X-ray spectrum as is the case for a bending magnet.

Radiation is emitted at each bend of the sinusoidal path. An observation point/detector located on the axis of radiation will receive a $2N$ photon flux, where $N$ is the number of periods of the sinusoid,
whereas the bending-magnet has only one bend and gives therefore much less intensity than the wiggler. For each magnetic period the intensity can be calculated by using Equation 2-22 or 2-23.

The principle of a wiggler is shown in Figure 2-7a. Here the oscillation of the electrons is shown with its X-rays emission at each individual bend where the electron momentum changes and $\alpha=k/\gamma >> 1/\gamma$. Furthermore, the cross section of the beam is smaller in horizontal size than in the case of a bending magnet.

**Undulator**

The undulator is the second type of insertion device which can be installed in a straight section of the storage ring. The path of the electrons in the undulator regime ($K=1$) is sinusoidal but with a smaller amplitude than that of a wiggler, see Figure 2-7b.

Due to the amplitude and period, the produced X-ray cones are forced to interfere with each other (Fig. 2-7b) and a peaked energy spectrum is obtained as is shown in Figure 2-8. This is in contrast to the X-ray cones of a wiggler regime which do not meet the conditions for interference. Furthermore, the intensity increases with $N^2$ and the horizontal cross section of the beam is smaller than that of a bending magnet.

**Figure 2-7: Principle of a wiggler (a) and an undulator (b) for their respective spectrum contribution (from Baruchel et al.).**

The undulator regime will not be discussed in more detail since an undulator source was not used in this experimental work.

Figure 2-8 shows the X-ray energy spectrum for a bending-magnet and the insertion devices.
Experimental stations

The X-ray beam is taken into a specially designed scientific station consisting of an optics and one or more experiments hutch, as is shown in Figure 2-4, containing the equipment necessary for the experiments as will be explained in more detail in the next chapters.

References


