Perturbed Angular Correlation with Synchrotron Radiation at the 67.4 keV Nuclear Resonance of ^{61}Ni

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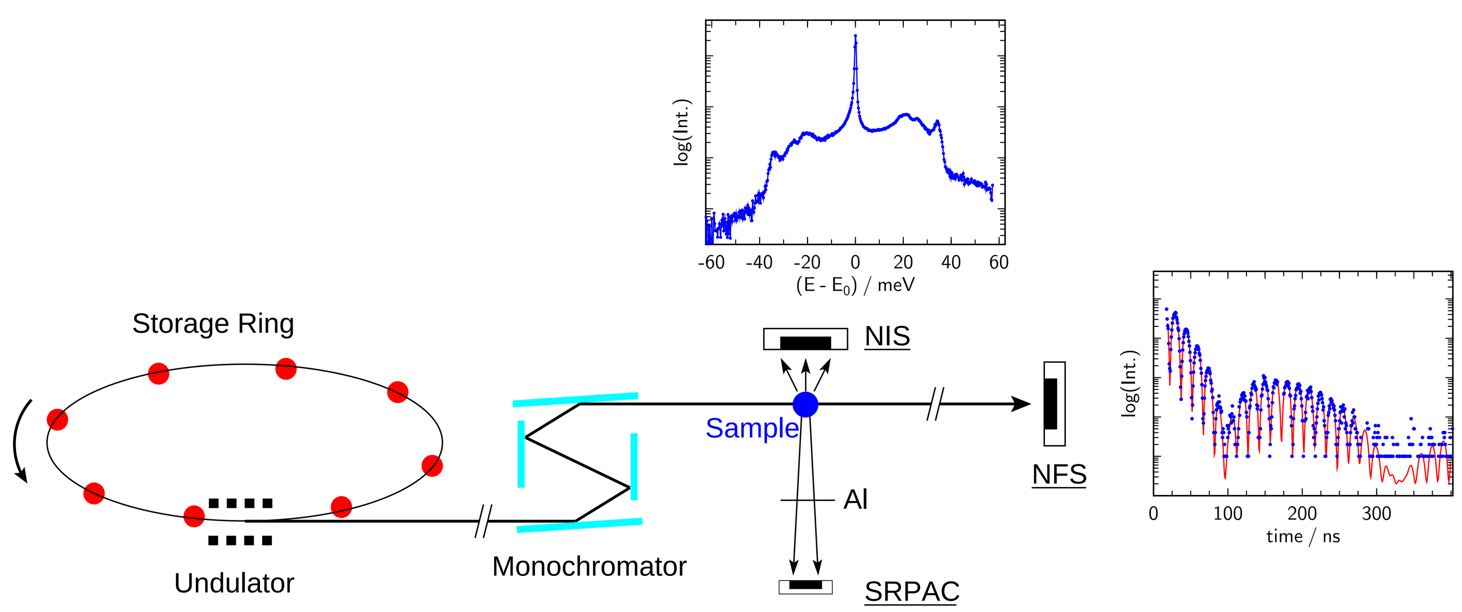
Introduction

The transition metal nickel is of great importance in nature and of high interest in many fields of research. Due to its magnetic properties, Ni is of particular importance in magnetism, both for magnetic applications and for basic research. Nickel plays also a very important role in biology, e.g., as active center in various metalloproteins.

Ni is rarely used in Mössbauer spectroscopy, due to the high γ-energy of 67.4 keV of the ^{61}Ni Mössbauer isotope, which implies a small Lamb-Mössbauer factor, and due the short life time of the parent nuclei.

In Nuclear resonant scattering (NRS) of synchrotron radiation (SR) the ^{61}Ni resonance is difficult to observe both due to the high γ-energy, which makes efficient high resolution monochromators merely impossible, and due to the short life time of the Mössbauer level.

The three main Branches of NRS



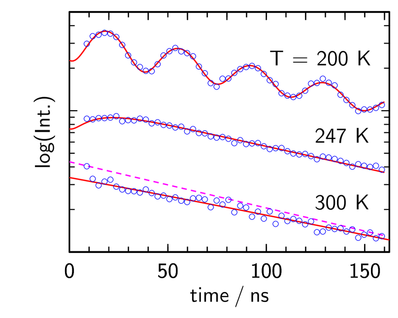


Figure 1: Schematic presentation of the three most important NRS techniques, exemplarily shown for the ^{57}Fe resonance. All three methods exploit the time structure of the SR.
In Nuclear Inelastic Scattering (NIS) the time integrated intensity following the pulsed excitation is recorded as a function of the energy of the exciting beam and yields information about dynamics in the sample.
Nuclear Forward Scattering (NFS) and SRPAC are time differential methods, the shape of the time decay of the intensity following the pulsed excitation of excited nuclear states gives information on the hyperfine fields acting on the nucleus.
The Al absorber in the SRPAC setup suppresses the X-ray fluorescence following the internal conversion process.

Experimental Method

Nuclear resonant scattering of synchrotron radiation at the ^{61}Ni resonance, which was first observed at the PETRA undulator station at HASYLAB, opens the way to this fascinating isotope with its numerous applications. We used a novel variant of NRS, namely Synchrotron Radiation based Perturbed Angular Correlation (SRPAC), the scattering analogue of the PAC-method. It is the virtue of this new technique that no radioactive parent isotope is needed, because the nuclear level is populated from the ground state via excitation by SR. The time modulation of the reemitted radiation reveals the hyperfine splitting of the excited nuclear state. Another advantage is that the scattering process is not perturbed by translational motions, e.g., diffusion, which destroys coherence in Nuclear Forward Scattering, and SRPAC can be performed "with recoil", i.e., at high temperatures and/or high gamma-energies, where the Lamb-Mssbauer-factor f_{LM} vanishes.

^{61}Ni nucleus with magnetic HFI

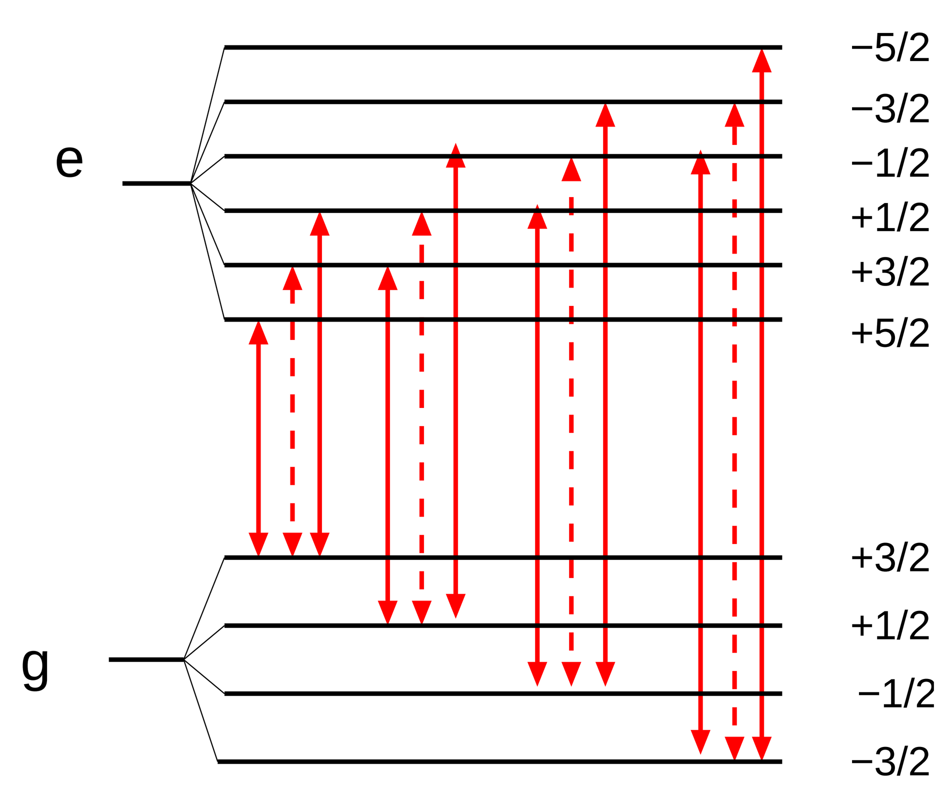


Figure 2: Hyperfine splitting of the nuclear levels of ^{61}Ni, with spins $I_g = 3/2$ and $I_e = 5/2$ of the ground and excited nuclear states, respectively. The magnetic Hyperfine Interaction (HFI) completely lifts the degeneracy of the nuclear states.
Arrows denote the possible γ-transitions for magnetic dipole (M1) radiation.
The full lines represent $\Delta m = \pm 1$ transitions, the dashed lines $\Delta m = 0$ transitions.

Comparison of NFS and SRPAC

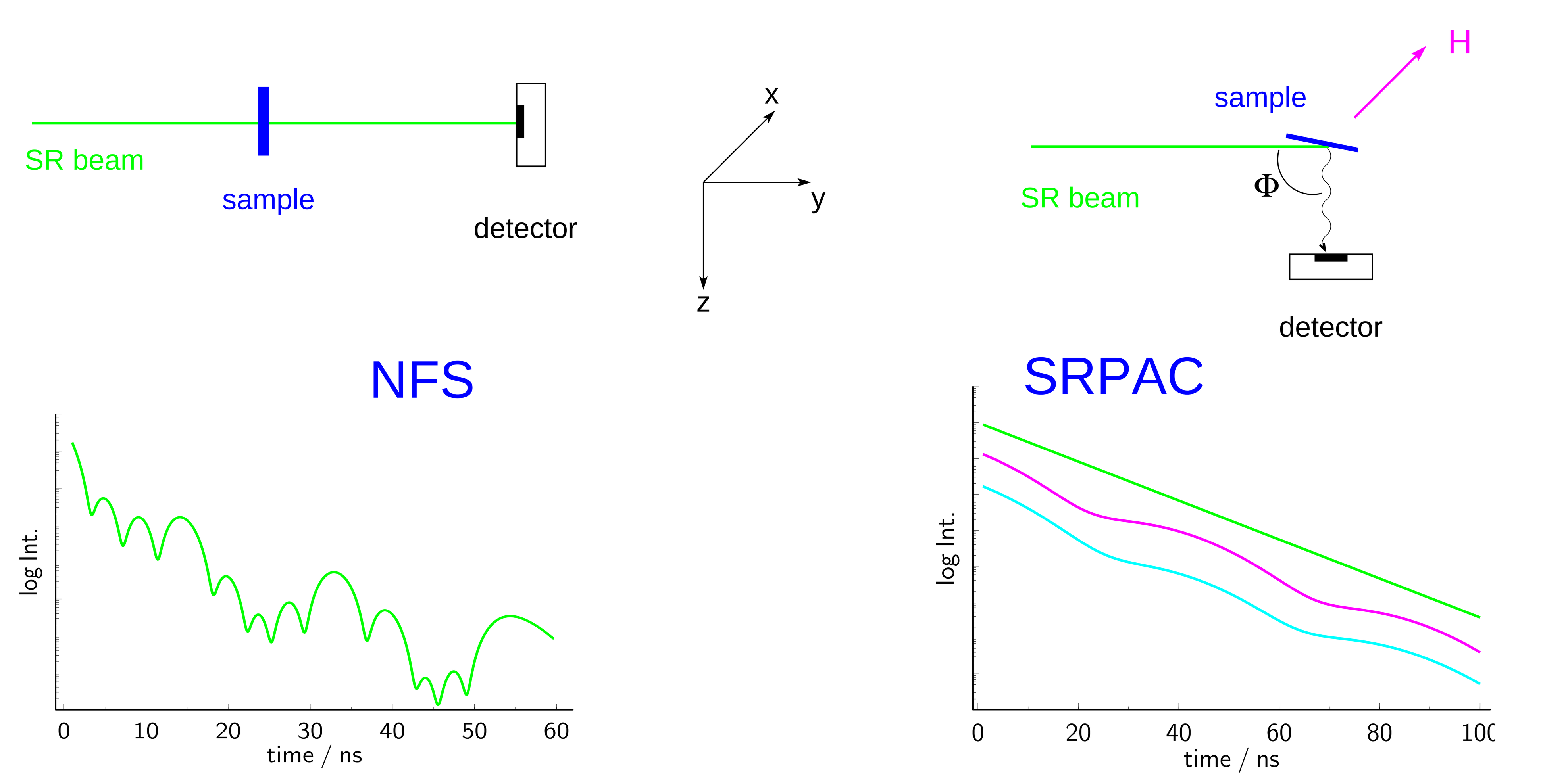


Figure 3a: NFS is a fully coherent scattering technique: all possible transitions are excited in phase by the SR pulse, the forward scattered intensity is a result of the coherent superposition of all scattering amplitudes from all nuclei in the sample.
The splitting of nuclear levels gives rise to quantum beats, if the magnetic HF field is randomly oriented, all transitions can be excited, there are 12 frequencies present in the time decay.
By appropriate orientation of the HF-field one can choose to excite either the $\Delta m = 0$ or the $\Delta m = \pm 1$ transitions (cf. Fig. 2).

Figure 3b: In contrast to NFS the SRPAC method is a single nucleus scattering technique. As a consequence, only the splitting of the excited state – here magnetic hyperfine (HF) splitting – gives rise to quantum beats.
If the magnetic HF field is randomly oriented, all transitions can be excited, there are two frequencies present in the time decay.
If the magnetic HF field is oriented in x- or y-direction, only the $\Delta m = 0$ transitions can be excited, there is only one frequency observed.
If the magnetic HF field is oriented in z-direction, there are no beats observable.

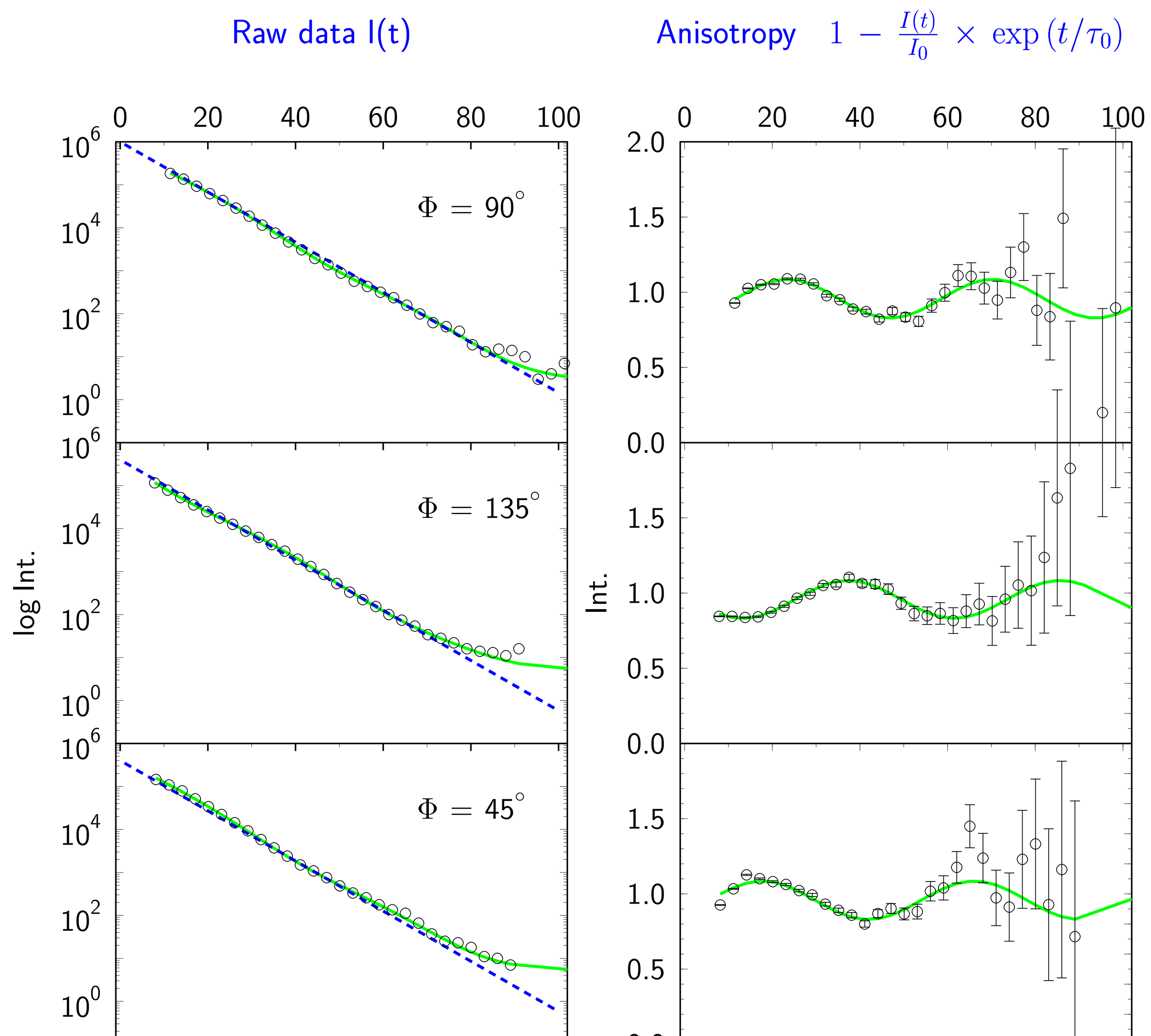


Figure 4: SRPAC time spectra recorded in an external magnetic field in the storage ring plane (x-direction, cf. Fig. 3b) at various observation directions of the reemitted γ-radiation (cf. Fig. 3b).
Black circles are the data points, the green full lines are fits with the function

$$I(t)=I_0 \times \exp(-t/\tau_0) \times (1-\frac{A}{2}(1+3\cos(2\Delta\Phi - 2\omega_B t)))$$

The dashed lines represent the unperturbed exponential decay with $\tau_0 = 7.4$ ns.
The quantum beats are clearly seen both in the raw data and in the so-called Anisotropy.
Note the phase shift for different observation angles.

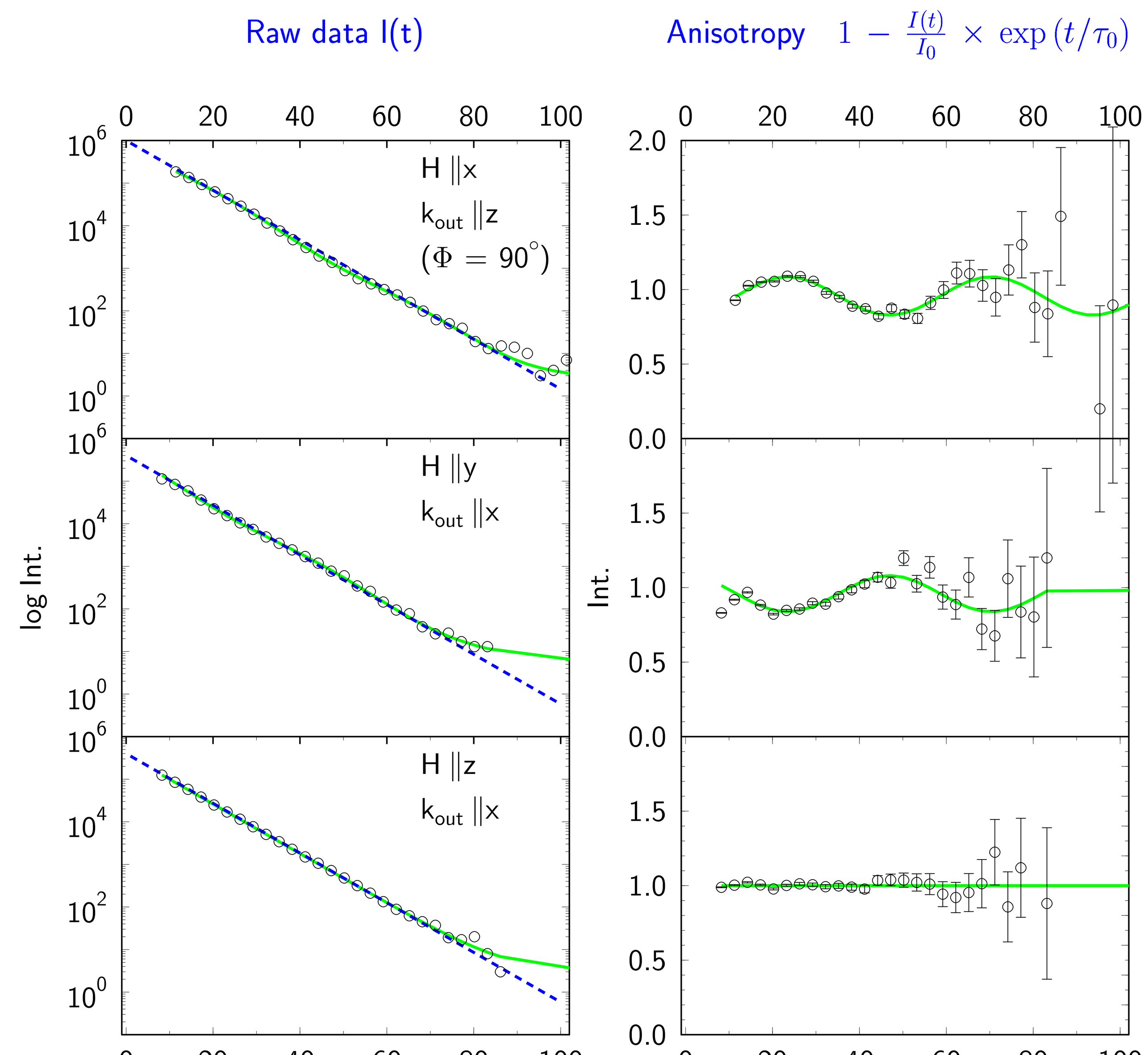


Figure 5: SRPAC time spectra recorded in an external magnetic field in various directions, and at various observation directions of the reemitted γ-radiation as indicated in the figure (notation of axes cf. Fig. 3b).
Black circles are the data points, the green full lines are fits. The dashed lines represent the unperturbed exponential decay.
bottom: the quantum beats disappear, when the magnetic field is perpendicular to the storage ring plane.
top and center: there is a phase change by 180° in the reemitted intensity when the field direction changes from x to y.

Summary

For the pioneering experiment of Synchrotron Radiation based Perturbed Angular Correlation we used an isotopically enriched ferromagnetic Ni-foil at room temperature in various scattering geometries with and without external magnetic field. The observed time modulation is in full agreement with theory. The technique will open new experimental possibilities so that a re-vitalization of PAC can be expected. Temperature dependent measurements on NiO and FeNi-invar alloy are in preparation.