Coherent Resonant X-ray Scattering from a Rotating Medium : The Nuclear Lighthouse Effect

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Introduction

Quantum objects like photons or neutrons acquire dynamical phase shifts upon propagation in rotating frames that can be measured in interferometry or diffraction experiments. The occurence of such interference effects requires the transit time of the particle through the system to be sufficiently long for the phase shifts to evolve. This condition can be fulfilled, for example, if a resonance with an energy width Γ_0 is excited in the system. The propagation of radiation is then affected by resonant scattering processes that take place on a time scale given by $\tau = \hbar/\Gamma_0$. If the resonance is excited by a radiation pulse that is short compared to τ, a collectively excited state (exciton) is created, where a single excitation is coherently distributed over the atoms of the sample. The coherence leads to remarkable properties of the exciton decay, notably the speedup of the radiative decay compared to that of an isolated atom, and quantum beats resulting from interference of waves emitted from different resonances at different atoms. The analysis of the temporal evolution of the subsequent radiative decay provides valuable information about the environment of the atoms in the sample. Therefore, this type of time-resolved spectroscopy has generated several applications in condensed matter physics, involving electronic and nuclear resonances [1,2]. In this contribution, we describe an effect that arises due to the evolution of dynamical phases upon *nuclear resonant scattering* in rotating frames.

Theory

Coherent resonant excitation of an ensemble of nuclei by x-rays leads to a bound state between the nuclei and the electromagnetic field, that is often referred to as 'nuclear exciton'. An interesting situation arises when a nuclear exciton is created in a rotating sample. If the collective phasing of the nuclei in the sample is preserved during the lifetime of the exciton, the exiton state is rotated with the sample. Consequently, the wavevector of the radiative decay $\vec{k}(t)$ deviates from $\vec{k}_0$ by the rotation angle φ that has developed during the time t after excitation. As a result, the time spectrum of the nuclear decay is mapped to an angular scale, as sketched in fig. 1. The time evolution of the exciton wavevector can be described by the precession equation : $\frac{d\vec{k}}{dt} = \vec{\Omega} \times \vec{k}$. With the initial condition $\vec{k}(0) = \vec{k}_0$, the solution is :

$$\vec{k}(t) = \vec{k}_0 - (\vec{k}_0 \times \vec{\Omega})\, t + \frac{1}{2}((\vec{k}_0 \times \vec{\Omega}) \times \vec{\Omega})\, t^2 - \dots \quad (1)$$

For the experiment to be discussed it is a good approximation to terminate this series after the first-order term. The wave resonantly scattered from the rotating sample is then given by :

$$\vec{A}(\vec{r}, t) = \exp[\, i\, (\vec{k}_0 - \vec{k}_0 \times \vec{\Omega}\, t) \cdot \vec{r}\,]\, \vec{A}_0(t) \quad (2)$$

where $\vec{A}_0(t)$ is the time response from the sample at rest which can be calculated within the framework of nuclear resonant forward scattering (NFS) [3].
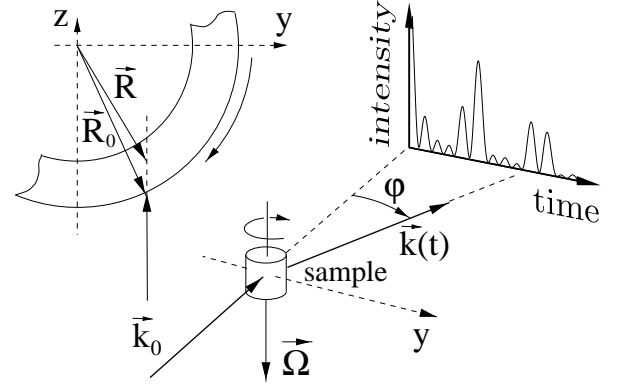

Figure 1: Scattering geometry of nuclear resonant scattering at a sample rotating with angular velocity $\vec{\Omega}$. The collectively excited nuclear state (nuclear exciton) is carried with the sample so that the time spectrum of the radiative nuclear decay is mapped to an angular scale [4].

Eq. 2 describes a wave that propagates in the direction of $\vec{k}(t) = \vec{k}_0 - \vec{k}_0 \times \vec{\Omega}\, t$. The mapping to an angular scale is described most conveniently by a one-dimensional transformation of eq. 2 into reciprocal space. Assuming fully coherent illumination, which is a good approximation under the present experimental conditions, the time response is given by $\vec{A}(\vec{q}, t) = \vec{A}_0(t)\, \delta(\vec{q} - \vec{k}_0 \times \vec{\Omega}\, t)$. Expressing the momentum transfer through $q = k_0\, \varphi$, integration over time t then yields the scattered intensity as a function of spatial coordinate only :

$$I(\varphi) = |\vec{A}_0(\varphi/\Omega)|^2 \quad (3)$$

This means that the angular distribution of the scattered radiation can be described by the theory of NFS using the mapping $t = \varphi/\Omega$. Due to the apparent analogy with a sweeping ray of light, this effect has been termed the *Nuclear Lighthouse Effect*. It was recently observed in nuclear resonant scattering from the 14.4 keV resonance of ^{57}Fe [4].

Experiment

For an experimental test of this prediction, we have employed nuclear resonant scattering at the 14.4 keV resonance of ^{57}Fe with a level width of $\Gamma_0 = 4.7$ neV and a lifetime of $\tau = 141$ ns. To obtain an angular separation of 0.1 mrad for events that are separated by 1 ns, rotational speeds in the order of 10 kHz are necessary. Such high-speed rotary motion is commonly used in the magic-angle-spinning (MAS) technique of solid-state NMR with frequencies up to 35 kHz.

The experiment was performed at the undulator beamline 3-ID at the Advanced Photon Source. The experimental setup is shown schematically in fig. 2.
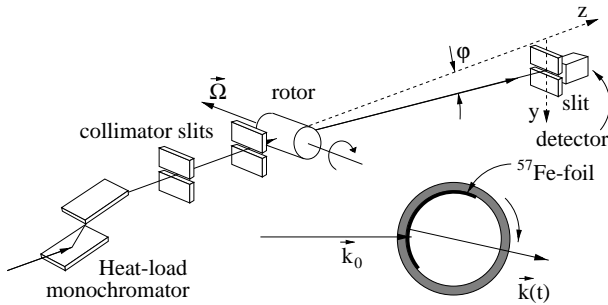

Figure 2: Scheme of the experimental setup [4]. The scattered radiation from the rotor was guided through an evacuated flight tube to the detector. The coordinates refer to the same frame as introduced in fig. 1. In the bottom right a cross sectional view of the scattering at the rotor is shown.

A slit system right after the heat-load monochromator was adjusted to define a beam cross section of $500 \times 30\,\mu m^2$ (horizontal $\times$ vertical), resulting in a typical flux of $2 \cdot 10^{10}$ s^{-1} on the sample. For sample rotation, a commercially available NMR - MAS system was used (Bruker Analytical GmbH, Germany), slightly modified for x-ray transmission. Samples were contained in hollow cylinders (4 mm diameter, 0.5 mm wall thickness) consisting of sintered polycrystalline Si_3N_4. In this experiment, the samples were spun around a horizontal axis with a rotational frequency of 18 kHz, corresponding to an angular velocity of 0.11 mrad/ns. After transmission through the rotor, the beam was guided through an evacuated flight tube to minimize small-angle scattering in air.

Results and Discussion

The angular distribution of the scattered radiation was recorded up to a scattering angle of 10 mrad relative to the primary beam, shown in fig. 3. Quantum beats of nuclear resonant scattering are clearly visible on top of a background (dotted line) that mainly results from small-angle scattering at the rotor material. The inset in fig. 3 shows the data after background subtraction. The solid line is a theoretical fit with the theory of NFS for a 5.7 μ thick Fe foil. This evaluation confirms that the time response of nuclear resonant scattering in rotating frames is mapped to an angular scale accord-
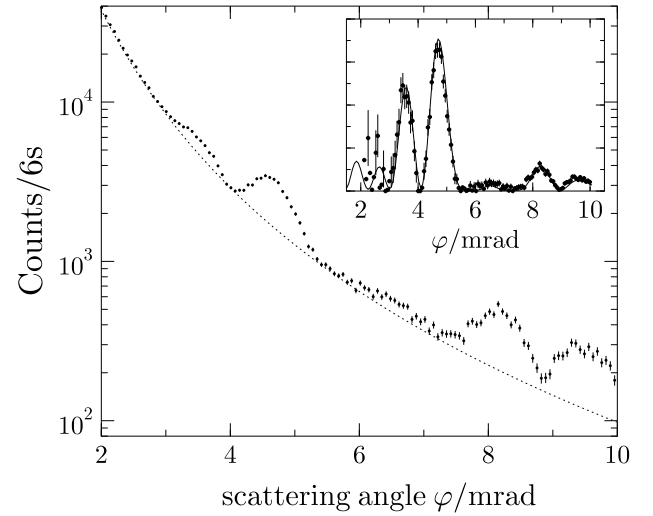

Figure 3: Measured angular distribution of 14.4 keV photons scattered from a 5.7 μm thick foil of ^{57}Fe spinning at 18 kHz [4]. The resonantly scattered quanta appear on top of the small-angle-scattering background that was measured independently and fitted with a power law (dotted line). The inset shows the background subtracted-data with a fit according to eq. 3 (solid line)

ing to eq. 3. The Nuclear Lighthouse Effect thus allows to obtain time spectra of nuclear resonant scattering by purely geometrical means. The temporal evolution of nuclear resonant scattering can be conveniently recorded as function of a spatial coordinate only, e.g., with an image plate. Since the magnitude of the angular deflection depends only on the time difference between excitation and reemission, this effect is independent of the time structure of the exciting radiation. As a result, this effect can be exploited in cases where the conventional timing technique is difficult, or impossible, to apply. For example, time resolutions can be achieved that are not possible with existing x-ray detectors and not even limited by the electron bunch length.

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