Experimental demonstration of time-integrated synchrotron-radiation spectroscopy with crossed polarizer and analyzer

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We present the results of a successful experiment performed with time-integrated synchrotron-radiation spectroscopy, a technique for studying hyperfine interactions. The measurement was performed on an enriched Fe sample in combination with a stainless-steel reference. A crossed polarizer/analyzer was used to reduce the prompt count rate. Time-integrated synchrotron-radiation spectroscopy is compared to the existing time differential synchrotron-radiation spectroscopy. The use of the polarizer/analyzer setup is discussed. Possible applications of time-integrated synchrotron-radiation spectroscopy are mentioned.

I. INTRODUCTION

The idea to use synchrotron radiation as a highly brilliant and polarized source to study nuclear resonances was launched in 1974 by Ruby.[1] It lasted until 1985 before Gerdau *et al.* made the unambiguous observation of synchrotron radiation resonantly scattered by ^{57}Fe nuclei.[2] Ever since, nuclear resonant fluorescence experiments with synchrotron radiation gained more and more interest as a useful means to study hyperfine interactions between resonant nuclei and their surrounding (see, e.g., Refs. 3–6 and many references therein). Measurements are generally performed in a time differential mode, in which the nuclear decay is studied as a function of time after excitation by the synchrotron burst. This time differential synchrotron-radiation spectroscopy (TDSRS) puts restrictions on the operation mode of the synchrotron, since the time interval between pulses should be comparable to the lifetime of the involved nuclear level, typically of the order of a few 100 ns (e.g., for ^{57}Fe: $\tau=141$ ns). A drawback of the method is the difficulty to study isomeric states with lifetimes of the order of μs (e.g., ^{181}Ta with $\tau=8.7$ μs), given the limited circumference of the storage ring. In 1996, we proposed a time-integrated technique[7] to do forward nuclear resonant scattering experiments with synchrotron radiation, yielding the same information on the hyperfine interactions, but easing the restrictions related to the nuclear lifetime. The idea is to perform time-integrated measurements by introducing a second, single-line reference sample that is Doppler modulated. Crossings between transition frequencies in the investigated sample and the variable reference frequency will show up as resonances in the forward scattered intensity.

Two experiments with this technique have been carried out on the 3ID undulator beamline at the advanced photon source (APS) in the Argonne National Laboratory. Details on this beamline can be found elsewhere.[8] The results of the first, preliminary experiment are described in Ref. 9 and show a high sensitivity to the applied time window and the dramatic effect of time slicing. In this paper we report on a second experiment, also performed at the same beamline, but in which we reduced the time slicing to a minimum by using a polarizer/analyzer setup.[10,11]

II. DESCRIPTION OF THE METHOD

The aim of time-integrated synchrotron-radiation spectroscopy (TISRS) is to perform measurements in energy domain. Since the incoming synchrotron radiation has a broadband spectrum $\Delta E \gg \Gamma$; Γ being the natural linewidth of the excited state), simply measuring as a function of energy of the incoming radiation is excluded. Hence, in addition to the investigated sample, we will introduce a single-line reference sample, which is mounted on a Mössbauer drive. Due to the velocity, the nuclear transition frequency will be Doppler modulated, according to $\omega_r=\omega_0(1+v/c)$, where v and c have the usual meaning of velocity and speed of light, ω_0 is the nuclear transition frequency in absence of any Doppler modulation, and ω_r is the modulated transition frequency in the reference. In the investigated sample, there can be several nuclear transitions, depending on the hyperfine interaction. This sample is kept stationary and, consequently, all frequencies will be constant.

Due to space coherence, synchrotron radiation can excite all nuclei in sample and absorber coherently. The coherence length is typically several meters. Moreover, the incoming synchrotron photon has a broadband spectrum and will excite all nuclear transition frequencies within its spectral band. This is well known as energy coherence. The coherent excitation of different energy levels gives rise to interferences in the scattered probability. When the scattered radiation is measured as a function of time, the interferences will show up as quantum beats with beat frequencies corresponding to the energy differences of all allowed transitions from all different nuclear sites.[12] However, if the time-averaged intensity is measured, the quantum beats will level out, except when the beat period becomes comparable to or larger than the nuclear lifetime of the excited state. This happens when

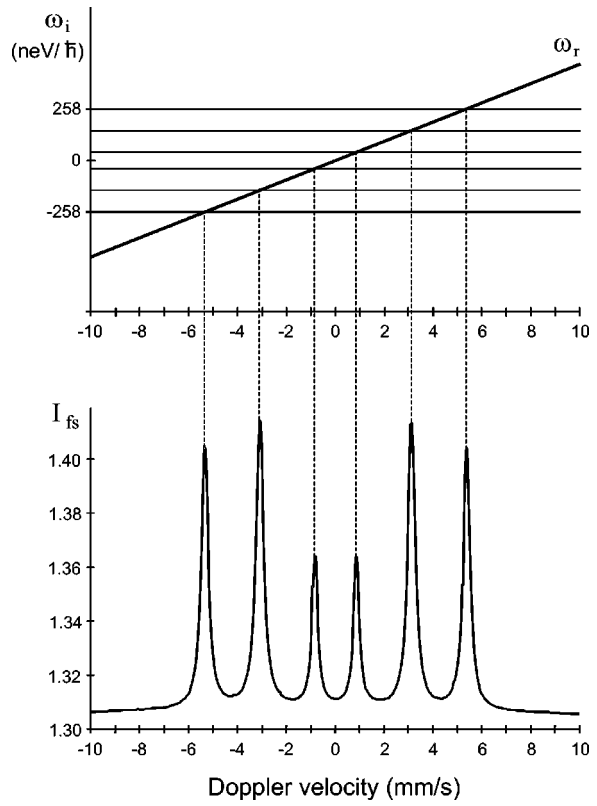

FIG. 1. Illustration of TISRS for a magnetically split Fe sample (magnetic field=33.3 T) and a single-line reference. Top: Transition frequencies in the sample (horizontal lines) and reference (inclined line) normalized to 14.413 keV. Below: The corresponding TISRS simulated spectrum showing forward scattered intensity versus velocity of the reference sample. (The effective thickness of the sample and reference are 5 and 1, respectively)

the reference frequency ω_r matches one of the transition frequencies in the sample. For thin samples, the forward scattered intensity is given by[9]

$$I_{fs}=N_s^2+N_r^2+\sum_i \frac{2N_sN_r}{(\omega_i-\omega_r)^2\tau^2+1}. \qquad (1)$$

N_s and N_r are proportional to the number of resonant nuclei in the sample and reference, respectively, and τ is the nuclear lifetime of the excited state. The first two terms, $N_s^2+N_r^2$, are constant. The summation in the third term runs over all nuclear transition frequencies ω_i in the investigated sample. Measuring the nuclear resonant forward scattered radiation as a function of the reference velocity yields a spectrum in energy domain. Whenever ω_r is equal to one of the ω_i, the last term in Eq. (1) will give an extra contribution, seen as a resonance. This is shown schematically in Fig. 1. The TISRS spectrum is very similar to an ordinary Mössbauer spectrum, and as a result, the interpretation is obvious.

Eq. (1) only takes into account nuclear scattering on sample and reference. In the configuration where also a polarizer and analyzer are present, the expression for the forward scattered intensity becomes

$$I_{fs}=\text{const}-\sum_i \frac{2N_sN_r\alpha_i}{(\omega_i-\omega_r)^2\tau^2+1}, \qquad (2)$$

where α_i is a polarization dependent factor, proper to the particular transition. The main difference with Eq. (1) is the minus sign before the summation, which results in negative resonances, i.e., the spectrum will show dips instead of peaks.[7] From the positions, the widths, and the amplitudes of the resonances, hyperfine and solid-state information can be obtained, similar as in classical Mössbauer spectroscopy. For thick samples Eqs. (1) and (2) look different, but the general features remain.[9,13]

III. EXPERIMENTAL DETAILS

An overview of the experimental setup is given in Fig. 2. The undulator is tuned to the 14.413 keV nuclear resonance of ^{57}Fe. With a high-heat-load Si(1 1 1) double crystal monochromator, an energy bandwidth of 3.5 eV at the nuclear resonance is selected. The flux right after the monochromator is 3×10^{13} photons/s. Since we want to detect radiation that is resonantly scattered in forward direction, the detector will see the full photon beam and will be saturated for several tens of ns after each synchrotron pulse. This complicates TISRS, because putting such a long time gate introduces serious distortions in the spectrum.[9] Therefore, it is crucial to reduce the nonresonant (prompt) intensity as much as possible. This is done by a crossed polarizer and analyzer.[11] These consist of two Si (8 4 0) channel-cut crystals ($\theta_B=45.1°$) with an asymmetry angle of $-43°$. The first crystal transmits the σ-polarized synchrotron radiation, while the second one, a similar crystal but turned over 90°, only transmits π-polarized light (see Fig. 2). So, only radiation that changes its polarization state from σ to π can reach the detector. This may occur for the resonant radiation when it is scattered on a hyperfine split sample. The nonresonant prompt radiation remains σ polarized and is reduced by the analyzer to an amount of 4×10^2 photons/s. This low value allows measuring from 1 or 2 ns after the pulse. Simulations have shown that such a small time gate has negligible effects on the measurement.

Two absorbers at room temperature are placed between the polarizer and the analyzer. The first absorber, the investigated sample, is a magnetically split Fe foil of 2.5 μm thickness, 53% enriched in ^{57}Fe. A small external magnetic field (0.27 ± 0.03 T) is applied to this sample in order to

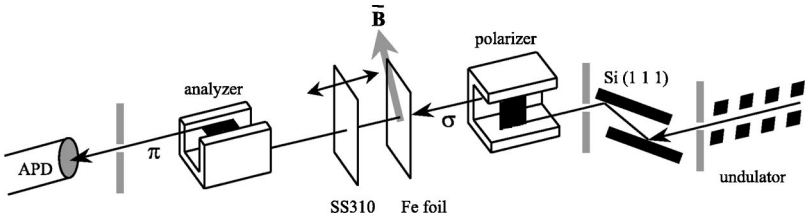

FIG. 2. Experimental setup. The propagation direction of the beam is from right to left.

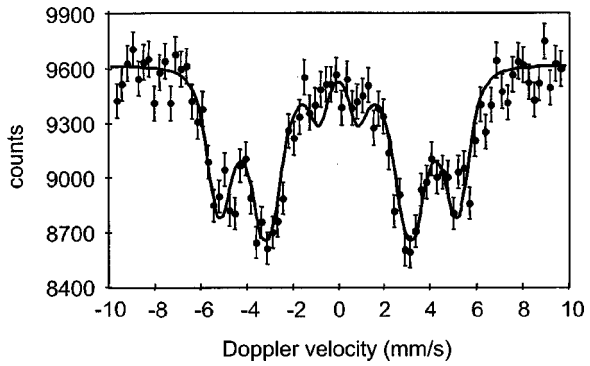

FIG. 3. TISRS spectrum of a magnetically split Fe sample (effective thickness=21) and a stainless-steel reference sample (effective thickness=18) in the presence of a crossed polarizer and analyzer. The solid line is the calculated theoretical spectrum, plus a constant background contribution.

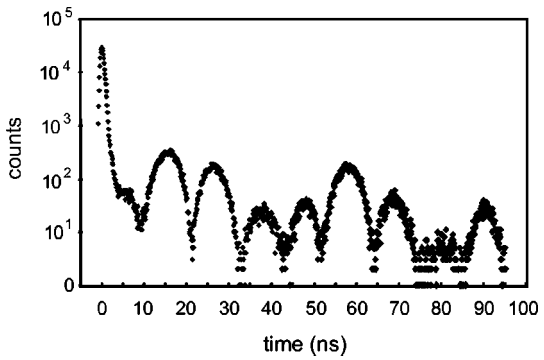

FIG. 4. TDSRS spectrum of a magnetically split Fe sample (effective thickness=21) in the presence of a crossed polarizer and analyzer.

orient the hyperfine field perpendicular to the beam direction and under an angle of 45° with respect to the linear polarization plane of the synchrotron. This particular direction of the magnetic field gives maximal transmission through the polarizer/analyzer setup. The second absorber is a 2.1 μm thick stainless-steel foil (SS310) with 95% ^{57}Fe enrichment. This one is mounted on a triangularly driven Mössbauer drive, and acts as a single-line reference sample.

The forward scattered photons are registered by a fast operating avalanche photodiode detector (20% efficiency) in intervals of 93 ns, starting from 2 ns after the synchrotron pulse. To guard against diffuse scattering from the walls, appropriate Pb shielding was done. The measurement was performed with a pulse separation of 100 ns and a mean resonant count rate of 40 photons/s. The data collection time was 5 h.

IV. RESULT

The resulting time integrated spectrum is given in Fig. 3, showing the forward scattered intensity versus the Doppler velocity of the reference sample. The smooth line is a simulation based on the semiclassical optical model for nuclear resonant scattering.[6] The spectrum clearly shows the six ^{57}Fe peaks corresponding to transitions involving an angular momentum change $\Delta m=0, \pm 1$. The relative intensity of the peaks is related to the degree of $\sigma \rightarrow \pi$ polarization change for that particular transition. The separation between the resonances provides information on the hyperfine interactions, while the resonance widths are related to the effective thickness of the samples.

For comparison, the TDSRS spectrum is given in Fig. 4. This spectrum is taken under the same conditions, after having removed the stainless-steel reference sample. The forward scattered intensity is now recorded versus time after excitation by the synchrotron pulse. The data collection time was 10 min with a resonant count rate of 55 photons/s. The beat pattern in the time dependent spectrum reflects all hyperfine information since the quantum beats are determined by the energy differences in the sample.[12,14]

V. DISCUSSION

Qualitative comparison between TISRS and TDSRS

TISRS and TDSRS yield the same hyperfine and solid-state information. Time differential synchrotron-radiation spectroscopy is obviously more suited in cases where time dependent phenomena are to be studied, or if one is particularly interested in the dynamics of the system. If, on the other hand, hyperfine interactions are the object of the measurement, then time-integrated synchrotron-radiation spectroscopy can be considered as well. It has the advantage over TDSRS to yield spectra that are easier to interpret. This becomes a real advantage when samples with many hyperfine field components are studied. The TISRS spectrum is just a superposition of spectra, each one belonging to a component. In TDSRS, on the other hand, all components interfere, so that each pair of transition frequencies produces a quantum beat. The number of quantum beats can become very large, resulting in a complicated beat structure.

An important difference between TISRS and TDSRS with crossed polarizer/analyzer concerns the data collection time. The spectra from Figs. 3 and 4 were recorded with comparable resonant count rates, yet for TISRS it took 30 times longer to get a good spectrum. This can be explained as follows. In case of TDSRS, the signal is formed by radiation resonantly scattered on the investigated sample. Since this radiation is the only one passing through the analyzer, one measures 100% signal. For TISRS, there are two processes that contribute to the spectrum. The first one, the most intense, is again radiation resonantly scattered by the investigated sample, without being scattered by the reference sample. This process has no velocity dependence and hence, it will only contribute to a constant background. The actual signal is formed by radiation that has been resonantly scattered both on the sample and on the reference. However, this is a double-scattering event. The extra scattering on the reference makes it — for the most intense line — a ten times less likely process than the single scattering on the sample only. As a consequence, the signal-to-background ratio is only 10%. In order to get comparable statistics as in TDSRS, one needs to measure ten times longer.

Additionally, there is another effect to account for. Data collection times will also be longer for TISRS due to the fact that counts are measured sequentially over the velocity scale. As in conventional Mössbauer spectroscopy, channels corre-

sponding to the baseline (i.e., off-resonance channels) yield no information on the hyperfine parameters. Since these channels are also considered, the statistics will be reduced to some extent. This effect accounts for the remaining factor three difference in data collection time. These two arguments make TISRS inferior to TDSRS from a statistical point of view.

Use of polarizer and analyzer in TISRS

Although the implementation of the polarizer and analyzer filters reduces the undesired prompt radiation, it limits the possible applications. In order to change the polarization state of the radiation, samples with a uniquely defined orientation of the hyperfine field are required. This is a minor disadvantage when purely magnetic systems are studied, but requires monocrystals if a quadrupole interaction is involved. Furthermore, the study of single-line samples with this setup is simply excluded.

Another drawback of using a polarizer and analyzer for TISRS is the low statistics. The resonant count rate was only 40 photons/s. This is two orders of magnitude smaller than in ordinary nuclear resonant scattering experiments, where a high-resolution monochromator is used to reduce the bandwidth of incoming radiation. This is an important factor when dealing with low resonant intensities. Data collection times of a few minutes (when no polarizer/analyzer is used) become several hours (with crossed polarizer/analyzer). If, however, the resonant intensity from the synchrotron can be increased by a factor of 100 or 1000, the data collection time will be a few seconds versus a few minutes. Then, in both cases, spectra with sufficient statistics are produced in a short time so that, although the relative difference in data collection time would still be two or three orders of magnitude, this difference is not relevant anymore and the setup with crossed polarizer/analyzer becomes interesting for TISRS measurements.

With currently available resonant intensities, however, the use of a polarizer and analyzer for time-integrated synchrotron-radiation spectroscopy is unfavorable from a statistical point of view. In order to make TISRS more generally applicable for the study of hyperfine interactions, one needs to find an appropriate way to filter out nonresonant radiation to an extent that is tolerable by the detector, without losing too many statistics. Therefore, other schemes for filtering out the nonresonant radiation have to be considered. In a previous publication,[9] the use of a nuclear monochromator has been proposed. The idea is to reduce the synchrotron-radiation bandwidth by Bragg scattering on a thick nuclear monochromator. A thorough analysis performed by Smirnov[15] has shown that in the case of ^{57}Fe, the energy bandpass of the nuclear monochromator should be 1–5 μeV. Only then, the following two requirements can be fulfilled. On the one hand, the delayed response of the monochromator should be short enough ($\leqslant 3$ ns), so that it can be gated out without distorting the TISRS spectrum. On the other hand, the transmitted intensity should be low enough ($\leqslant 10^6$ cps) not to saturate the detector. However, presently available nuclear monochromators have a bandpass of less than 0.5 μeV,[16–19] corresponding to a delayed response of ± 30 ns, which is intolerable for TISRS. How this bandpass could be increased by one order of magnitude is not evident. Other possible solutions, such as the realization of an ultrafast shutter[15] or the development of a string of ultrathin detectors,[20] still need to be investigated.

Possible applications of TISRS

TISRS can be a useful technique to study hyperfine interactions, especially in cases where time-differential synchrotron-radiation spectroscopy and classical Mössbauer spectroscopy are not straightforward. For example, the study of ^{181}Ta with TDSRS is extremely difficult because of the long lifetime of the excited state ($\tau = 8.73$ μs). Measuring a time spectrum, as was done by Chumakov et al.,[21] is only possible under extreme conditions: the storage ring should be operated in a single bunch timing mode. But even then, the time interval between pulses was only 780 ns,[21] which is not even one tenth of the natural nuclear lifetime. As a consequence, one needs samples with a large effective thickness so that the effective nuclear lifetime of the excited state is sufficiently decreased due to speedup[22] and the nuclear decay can be measured during the time window. For this isotope, TISRS is a good alternative. It can be performed in a more efficient timing mode (a pulse separation of ± 20 ns) and will provide reliable hyperfine information. Another example is ^{73}Ge. It also has a long lifetime of the excited state ($\tau = 4.26$ μs), which complicates TDSRS for the study of hyperfine interactions. On the other hand, classical Mössbauer experiments are not possible because there is no source available. Again, time-integrated synchrotron-radiation spectroscopy could be a solution.

The most interesting application of TISRS, however, can be found in the study of very small hyperfine splittings in nuclei with a long lifetime of the excited state. If the hyperfine splittings are less than 0.1 neV, the corresponding quantum beat period becomes larger than 2 μs. Time windows available at the present synchrotron-radiation storage rings are too short to follow such a slow quantum beat pattern, and hence, most hyperfine information is not accessible for time differential synchrotron-radiation spectroscopy. With the time integrated method, on the other hand, these hyperfine splittings can be observed if the linewidth is small so that the different resonance lines in the spectrum are still resolved.

VI. CONCLUSION

With this experiment, we were able to prove experimentally the feasibility and usefulness of TISRS. The main advantage is that it is less dependent on the time mode of the synchrotron machine. If in the future the intensity of synchrotron radiation at storage rings will be increased, then the full power of TISRS will be obvious: it is a complementary method to TDSRS and classical Mössbauer spectroscopy. In some cases where these two methods become difficult or even impossible, TISRS provides a useful solution. Typical examples are ^{181}Ta and ^{73}Ge where time-integrated synchrotron-radiation spectroscopy would be a good alternative.

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