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Coherent nuclear resonant scattering by ^{61}Ni using the nuclear lighthouse effect

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We have observed coherent nuclear resonant scattering of synchrotron radiation from the 67.41-keV level of ^{61}Ni. The time evolution of the forward scattering signal was recorded by employing the nuclear lighthouse effect. This method is used to investigate Mössbauer isotopes in a coherent scattering process with synchrotron radiation at high transition energies. The decay of the excited ensemble of nuclei in Ni metal shows quantum beats that allowed the determination of the magnetic hyperfine field at the ^{61}Ni nucleus. Moreover, we determined the lifetime of the 67.41-keV level of ^{61}Ni to be 7.4(1) ns.

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Over the last two decades, nuclear resonant scattering (NRS) of synchrotron radiation (SR) experienced a huge success in complementing the use of radioactive sources needed for traditional Mössbauer spectroscopy.[1] Like Mössbauer spectroscopy, nuclear forward scattering[2] (NFS) is an excellent tool to examine hyperfine interactions in solids.[3] Thanks to the brilliance of present SR sources, experiments with very small sample sizes such as under high pressure[4] or ultra-thin films in grazing incidence[5] became feasible. Furthermore, new concepts evolved that had no counterpart in traditional Mössbauer spectroscopy, such as nuclear inelastic scattering[6,7] or synchrotron-radiation-based perturbed angular correlation.[8,9]

Mössbauer transitions are characterized by a very narrow energy width in the neV region. NRS of SR has been restricted to a few isotopes, where sophisticated high-resolution Bragg monochromators allowed the reduction of the large electronic scattering background by decreasing the large SR bandwidth down to the meV region.[10,11] Only then, electronic gating of the signal from fast detectors[12,13] permits the discrimination between the rapid electronic scattering and the delayed response of the decaying excited nuclear level. Unfortunately, high-resolution monochromators of the "nested" type[10] are restricted to an energy range of up to approximately 30 keV. The reason for this is the decreasing angular acceptance of the Bragg reflection as x-ray energies increase. This is less an issue for Bragg backscattering.[11,14] Nonetheless, above 50 keV, this method seems extremely challenging: Very thick perfect crystals are needed, as the x-ray extinction depth is in the centimeter range, further requiring an enormous temperature stability, and the reflectivity is only a few percent.

Apart from experiments with the 14.41-keV transition of ^{57}Fe, which is the most often used isotope, experiments with ^{169}Tm (8.41 keV), ^{119}Sn (23.88 keV), ^{83}Kr (9.4 keV), ^{181}Ta (6.21 keV), ^{151}Eu (21.54 keV), ^{161}Dy (25.65 keV) (Ref. 15), ^{149}Sm (22.49 keV) (Ref. 4), ^{129}I (27.75 keV) (Ref. 16), and ^{40}K (29.83 keV) (Ref. 17) have been performed. Very recently, the method of backscattering monochromatization allowed investigations with ^{121}Sb (37.13 keV).[18] Other isotopes with high-transition energies have only been excited and their fluorescence decay could be detected. Spectroscopic work has not been performed, due to the problems mentioned above. This is the case for ^{61}Ni (67.41 keV) (Ref. 19) and ^{197}Au (77.35 keV) (Ref. 20).

Lately, a method was described that allows a modified NFS experiment that does not require the use of high-resolution monochromators: the nuclear lighthouse effect[21] (NLE). When the sample is subjected to a fast rotation, the time evolution of the decaying nuclei is translated into an angular deviation: With increasing time after excitation, the reemitted photons are deflected off axis with increasing angular deviation α from the forward direction. This method has been applied to ^{57}Fe,[21] ^{119}Sn,[22] and ^{149}Sm.[23]

Here, we have studied the hyperfine interaction of ^{61}Ni with the NLE. Nickel is a very interesting system in many aspects. Like other 3*d* transition elements, it exists in different valence states, shows a multitude of striking magnetic properties such as in iron-nickel Invar alloys,[24] and plays a key role in biomolecules.[25] It is therefore of great interest for hyperfine interaction studies. Traditional Mössbauer spectroscopy with this isotope[26,27] is very much hampered by the short lifetimes of the two possible mother isotopes ^{61}Co and ^{61}Cu, which have lifetimes of 99 and 213 min, respectively.

The experiments were performed in the 16-bunch filling mode at the nuclear resonance beamline ID18 of the European Synchrotron Radiation Facility.[28] Although the NLE does principally not depend on the type of filling mode,[21] additional time discrimination was necessary in the case of ^{61}Ni: The nuclear signal of ^{61}Ni is quite small as compared to the electronic scattering background mainly caused by small-angle x-ray scattering (SAXS). As the latter is scattering on a "prompt" time scale of 10^{-15} s, the 16-bunch mode together with a fast detector permits a time discrimination between the two processes. The reason for the small ratio of the nuclear signal to the electronic scattering is the low Lamb-Mössbauer factor f_{LM} for high-energy nuclear transitions.[29]

The setup for nuclear lighthouse effect experiments is shown in Fig. 1. The key part is a modification of a commercially available rotor/stator system[30] conceived for magic angle spinning NMR. The modifications allow for x-ray transmission and cooling down to 100 K, although rotor instabilities limit us to around 150 K so far. Gas flows assure

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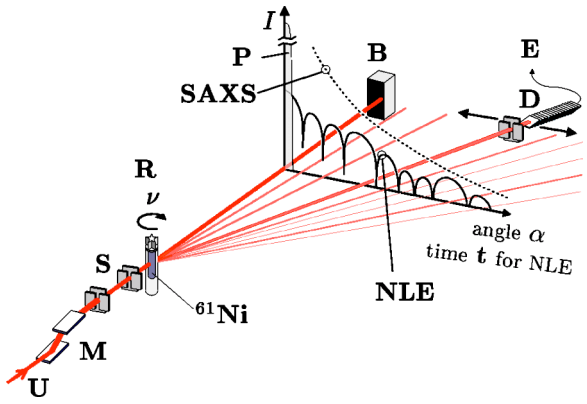

FIG. 1. (Color online) Experimental setup. The undulator radiation U is monochromatized coarsely ($\delta E=9$ eV) by two Si (3 3 3) reflections M and shaped by a slit system S. The rotor R containing ^{61}Ni is spinning at $\nu=8$ kHz. The noninteracting prompt direct beam (intensity P) hits the beam stop B. The prompt small-angle x-ray scattering SAXS is in competition with the delayed nuclear lighthouse effect signal NLE, thus demanding fast detection with an array of avalanche photo diodes D and time-discriminating electronics E. The detector scans over a range of angles. Only for the nuclear delayed signal, angles are resolved proportional to the interaction time.

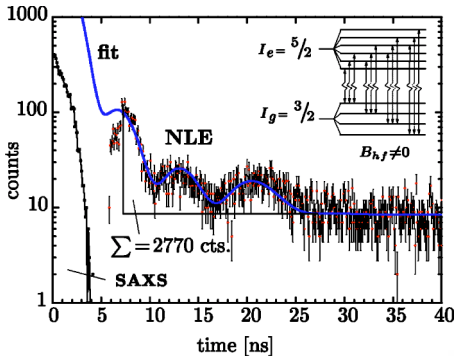

FIG. 2. (Color online) The nuclear lighthouse effect with ^{61}Ni at 180 K. The detector signal was gated off during the first 7 ns to discriminate the prompt SAXS background (~10 kHz). The SAXS is seen at early times when removing the gate temporarily for a short instant. The NLE data could be fitted by the theory of nuclear forward scattering using the program MOTIF (Ref. 33). The beat structure originates from the interference of the 12 transitions between the hyperfine-split ground and the 67.41-keV excited states in the presence of a magnetic hyperfine field B_{hf}, as illustrated on the top right. Without hyperfine interactions, the magnetic sublevels would be degenerate.

acceleration and bearing. The cooling is performed with an additional gas flow passing through a liquid nitrogen bath heat exchanger and directed to the central part of the rotor. PT100 sensors measure the gas temperatures. Important sealings were made of indium joints. A casing of two invar metal plates connected with invar screws and spring washers has been used to enclose the stator body tightly.

The graph in the center of Fig. 1 illustrates the photon intensities from three different processes. The huge peak P to the left corresponds to noninteracting undeviated photons and thus defines the time $t=0$. Compton scattering and SAXS contribute to deviated photons, but their interaction time with the electrons of the sample and the rotor walls is on a prompt time scale of around 10^{-15} s. This part is sketched by a dotted line following an angular dependence of α^{-4} according to Porod's law for SAXS from randomly oriented particles.[31] The third category are the photons that are scattered via excitation of the 67.41-keV nuclear level in ^{61}Ni. They are both delayed and deviated to the side according to the time spent in the foil before deexcitation. In favorable cases such as at the ^{57}Fe resonance, both the SAXS and NRS signals are of comparable strengths. The discrimination can be done with successive measurements of the angular intensity variation, once at resonance, once slightly detuned, and subtraction.[21] In the case of ^{61}Ni this is no more possible and fast detectors are employed to use the time information to discriminate between SAXS and the nuclear signal. Without discriminating detector electronics, the slit size in front of the detector defines the time resolution and each detector position $x=d\alpha$ corresponds to a time t according to $x=2\pi d\nu t$, with d being the distance between sample and detector, and ν the rotational frequency of the rotor. $x=0$ is the position of the direct beam. If, instead, time discrimination is applied, the time resolution is given by the detector and its electronics. Then, a large detector is best if it can bear the overall incoming flux. With small detectors as in our setup, which do not cover the whole angular region of interest, one needs to scan uniformly over the whole angular region. It should be stressed again that using fast detectors and time-discriminating electronics alone in standard NFS geometry without a viable high-resolution monochromator fails. The prompt flash of noninteracting photons hitting the detector is too intense to detect the smaller nuclear resonant signal shortly afterwards. Thanks to the NLE, the prompt undeviated beam and the delayed nuclear signal are separated in space.

The sample consisted of two isotopically enriched ^{61}Ni foils (85% enrichment), each 20-μm thick. Each foil was rolled to a tube that fitted tightly into a hollow ZrO_2 rotor of 3-mm inner diameter. The outer rotor diameter is 4 mm. The ZrO_2 rotor absorbs 70% of the 67.41-keV photons and gives rise to some SAXS. The undulator radiation, having 1/3 of that energy (22.47 keV), is transmitted by the Si (1 1 1) reflection of the monochromator, but it is completely absorbed by the ZrO_2 rotor walls. Therefore, it does not contribute to the SAXS background. Single crystalline sapphire rotors, as used in previous experiments[32] with ^{57}Fe samples, would absorb less and give less SAXS for the 67.41-keV radition, but are brittle and can break in the spinning unit at low temperatures.

The measurements at 180 K are presented in Fig. 2. We used a stack of 24 silicon avalanche photo diodes, each 100-μm thick and 10×10 mm^2 large, and inclined to an angle of 37° between the silicon surface and the beam direction. A detector slit limited the sensitive area to 6×4 mm^2, horizontal$\times$vertical. At 67.41 keV, the calculated efficiency of the total assembly is 20%. The electronic time resolution was 2 ns. During 375 min, the detector array was scanned repeatedly over a lateral displacement x ranging from 0 (be-

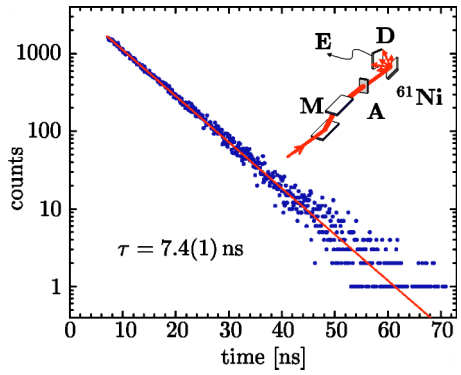

FIG. 3. (Color online)Time evolution of the fluorescence radiation of the 67.41-keV ^{61}Ni level. The setup to measure the nuclear fluorescence consists of two Si monochromator crystals *M* on the (3 3 3) reflection, an absorber *A* to reduce photon intensity transmitted via the Si (1 1 1) reflection, the ^{61}Ni foil, and a fast detector *D*, with time-discriminating electronics *E*. An electronic gate blocks data aquisition during the first 7 ns, i.e., especially the prompt photons.

hind the beam stop) to 50 mm. With ν=8 kHz and d =13.9 m, the corresponding times are 0 to 72 ns. Between two scans, the monochromator energy was checked using a setup as depicted in the inset of Fig. 3. The time discrimination in order to block the prompt signal of the SAXS background is done with an electronic gate that opens 7 ns after the arriving SR pulse. A multichannel analyzer (MCA) records the time spectrum (10 channels per nanosecond; 400 channels are displayed in Fig. 2). The detector is 6 mm wide laterally, corresponding to a temporal width of 8.6 ns. This defines the time window where photon counting is possible at each detector position. The precise time information within this window is obtained with the timing electronics and the MCA. The time resolution is thus given by the detector with its electronics and not by the detector slit size. The overall time the detector was in place to acquire delayed photons for a particular data point in Fig. 2 was 45 min. The average beam current in the storage ring was 70 mA. We detected 2770 photons being scattered by the ^{61}Ni nuclei, after subtracting the background solely due to cosmic radiation or electronic noise. This corresponds to a count rate of 1 s^{-1}, if we had a large detector covering the whole interesting angular range at once. The rate of incoming photons in the energetic width of the resonance is 300 s^{-1} at the stated electron beam current.

We could fit the data using the program MOTIF (Ref. 33) and obtain a magnetic hyperfine field of 6.7(2) T and an additional line broadening of 0.45 natural linewidths Γ_0. As input parameters, we use μ_g=−0.75 μ_N and μ_e=0.48 μ_N for the nuclear magnetic moments of ^{61}Ni (Ref. 34) and f_{LM} =0.025 according to the Debye model with a Debye temperature of 390 K.[35] The lifetime of the excited level was taken to be 7.4 ns, as determined by us and explained below. The magnetic hyperfine field at this temperature as measured with NMR is 7.2 T.[36] This slight discrepancy might be due to a small relative movement between the two sides of the cylindrical foils as seen by the beam. This lateral movement can be due to a precession of the rotor and might cause a Doppler shift acting on the transition energies.[37] An inhomogeneous line broadening was also observed in the NMR measurements[36] and attributed to impurities and strain.

For an independent determination of the lifetime of the 67.41-keV level, we measured the temporal evolution of the incoherent scattering of individual ^{61}Ni nuclei (nuclear fluorescence). The coherence is destroyed, when phonons intervene and an energy transfer (phonon creation or annihilation) takes place during the nuclear transition. The decay of individual excited nuclei follows an exponential behavior and the reemitted photons proceed into 4π as opposed to the forward direction in the case of NFS and the NLE. Some precautions have to be taken when realizing the setup shown in the inset of Fig. 3. The ^{61}Ni metal foil was kept at room temperature to obtain a negligibly low Lamb-Mössbauer factor. Otherwise, coherent effects like a propagating delayed wave field through the sample due to NFS (Ref. 38) would influence the exponential decay. Moreover, by covering a large solid angle with the detector, effects from the angular correlation between incident and scattered photons affecting the temporal evolution are averaged out.

The measured exponential decay is shown in Fig. 3. The detector was gated during the first 7 ns. The lifetime of the 67.41-keV level of ^{61}Ni was determined to be 7.4(1) ns. The adopted literature value is 7.70(23) ns, based on two measurements.[39] Shipley *et al.* report a value of 7.3(3) ns (Ref. 40), which is closer to our result.

In the future, other isotopes in this energy region could be addressed as well, especially ^{237}Np (59.54 keV and τ =97 ns), ^{193}Ir (73.04 keV, τ=8.8 ns), but also ^{189}Os (69.53 keV, τ=2.3 ns) and ^{73}Ge (68.75 keV, τ=2.5 ns). To increase the signal-to-noise ratio, several improvements are possible. First, a collimating compound refractive lens[41] (CRL) can increase the count rate by a factor of 10: The vertical divergence of the photons from the undulator is σ =12 μrad full width at half maximum. It can be decreased by a 80-hole parabolic aluminium CRL to $\Delta\theta$=0.9 μrad, which matches the acceptance of a 1° asymmetrically cut Si (3 3 3) reflection. Second, a further improvement of the cooling scheme to reach standard operation at 100 K without rotor stability problems seems reachable. The lateral rotor movement or a precession of the rotor can be disregarded, if only 1/3 circumference sample foils are used, as the beam passes the sample only once. Lower temperatures increase tremendously the Lamb-Mössbauer factor and operation at 100 K will give about another factor of 10 higher NLE signal. If the stability problems are solved, the use of sapphire rotors would be beneficial in decreasing both the SAXS signal and the absorption of 70% of the 67.41-keV photons. Rotational frequencies can still increase to 12 kHz. Finally, although linear detectors with high efficiency at these energies are missing, detector improvements can give at least a factor of 2, e.g., with two sets of detectors that scan half of the angular range simultaneously.

In conclusion, we have performed coherent nuclear resonant scattering from the 67.41-keV level of ^{61}Ni by

employing the nuclear lighthouse effect. We found a magnetic hyperfine field of 6.7(2) T at 180 K. The experiment opens the field for hyperfine spectroscopy at high-transition energies. The method of the nuclear lighthouse effect is extended to lower temperature conditions and might be applied to temperature-dependent phase transitions in the future.

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[1] E. Gerdau, R. Rüffer, H. Winkler, W. Tolksdorf, C. P. Klages, and J. P. Hannon, Phys. Rev. Lett. **54**, 835 (1985).
[2] J. B. Hastings, D. P. Siddons, U. van Bürck, R. Hollatz, and U. Bergmann, Phys. Rev. Lett. **66**, 770 (1991).
[3] O. Leupold, Hyperfine Interact. **144/145**, 21 (2002).
[4] A. Barla, J. P. Sanchez, Y. Haga, G. Lapertot, B. P. Doyle, O. Leupold, R. Rüffer, M. M. Abd-Elmeguid, R. Lengsdorf, and J. Flouquet, Phys. Rev. Lett. **92**, 066401 (2004).
[5] R. Röhlsberger, T. Klein, K. Schlage, O. Leupold, and R. Rüffer, Phys. Rev. B **69**, 235412 (2004).
[6] M. Seto, Y. Yoda, S. Kikuta, X. W. Zhang, and M. Ando, Phys. Rev. Lett. **74**, 3828 (1995).
[7] E. E. Alp, W. Sturhahn, T. S. Toellner, J. Zhao, M. Hu, and D. E. Brown, Hyperfine Interact. **144/145**, 3 (2002).
[8] A. Q. R. Baron, A. I. Chumakov, R. Rüffer, H. Grünsteudel, H. F. Grünsteudel, and O. Leupold, Europhys. Lett. **34**, 331 (1996).
[9] I. Sergueev, Ph.D. thesis, Technische Universität München, 2004.
[10] T. S. Toellner, Hyperfine Interact. **125**, 3 (2000).
[11] Y. V. Shvyd'ko, *X-Ray Optics* (Springer, Berlin, 2004).
[12] S. Kishimoto, Rev. Sci. Instrum. **63**, 824 (1992).
[13] A. Q. R. Baron, Hyperfine Interact. **125**, 29 (2000).
[14] Y. V. Shvyd'ko and E. Gerdau, Hyperfine Interact. **123/124**, 741 (1999).
[15] For an overview, see O. Leupold A. I. Chumakov, E. E. Alp, W. Sturhahn, and A. Q. R. Baron, Hyperfine Interact. **123/124**, 611 (1999).
[16] S. Kitao (private communication).
[17] M. Seto, S. Kitao, Y. Kobayashi, R. Haruki, T. Mitsui, Y. Yoda, X. W. Zhang, S. Kishimoto, and Y. Maeda, Hyperfine Interact. **141**, 99 (2002).
[18] H.-C. Wille *et al.* (unpublished).
[19] H.-C. Wille, M. Gerken, E. Gerdau, Y. V. Shvyd'ko, H. D. Rüter, and H. Franz, Hyperfine Interact. **C5**, 1 (2002).
[20] S. Kishimoto, Y. Yoda, M. Seto, S. Kitao, R. Haruki, T. Kawauchi, K. Fukutani, and T. Okano, Phys. Rev. Lett. **85**, 1831 (2000).
[21] R. Röhlsberger, T. S. Toellner, W. Sturhahn, K. W. Quast, E. E. Alp, A. Bernhard, E. Burkel, O. Leupold, and E. Gerdau, Phys. Rev. Lett. **84**, 1007 (2000).
[22] R. Röhlsberger, T. S. Toellner, K. W. Quast, W. Sturhahn, E. E. Alp, and E. Burkel, Nucl. Instrum. Methods Phys. Res. A **467**, 1473 (2001).
[23] R. Röhlsberger, K. W. Quast, T. S. Toellner, P. L. Lee, W. Sturhahn, E. E. Alp, and E. Burkel, Phys. Rev. Lett. **87**, 047601 (2001).
[24] M. van Schilfgaarde, I. A. Abrikosov, and B. Johansson, Nature (London) **400**, 46 (1999).
[25] R. P. Hausinger, JBIC, J. Biol. Inorg. Chem. **2**, 279 (1997).
[26] F. E. Obenshain and H. H. F. Wegener, Phys. Rev. **121**, 1344 (1961).
[27] P. Gütlich, R. Link, and A. Trautwein, *Mössbauer Spectroscopy and Transition Metal Chemistry* (Springer, Heidelberg, 1978), pp. 113–124.
[28] R. Rüffer and A. I. Chumakov, Hyperfine Interact. **97/98**, 589 (1996).
[29] The Lamb-Mössbauer factor in the Debye model is

$$f_{\mathrm{LM}} = \exp\left(- \frac{3E_\gamma}{m_{\mathrm{Ni}} c^2 k_B \Theta_D} \left[\frac{1}{4} + \frac{T^2}{\Theta_D^2} \int_0^{\Theta_D/T} \frac{y}{e^y - 1} \mathrm{d}y \right] \right),$$

with E_γ being the transition energy.
[30] Bruker Analytik GmbH, Rheinstetten, Germany.
[31] G. Porod, in *Small Angle X-ray Scattering*, edited by O. Glatter and O. Kratky (Academic Press, London, 1982), pp. 17–51.
[32] R. Röhlsberger, K. W. Quast, T. S. Toellner, P. L. Lee, W. Sturhahn, E. E. Alp, and E. Burkel, Appl. Phys. Lett. **78**, 2970 (2001).
[33] Y. V. Shvyd'ko, Phys. Rev. B **59**, 9132 (1999).
[34] *Table of Isotopes*, 8th ed., edited by R. B. Firestone and V. S. Shirley (Wiley, New York, 1996), p. 283.
[35] R. J. Birgenau, J. Cordes, and A. D. B. Woods, Phys. Rev. **136**, A1359 (1964).
[36] R. L. Streever and L. H. Bennett, Phys. Rev. **131**, 2000 (1963).
[37] K. Quast, Ph.D. thesis, Universität Rostock, 2002.
[38] U. Bergmann, J. B. Hastings, and D. P. Siddons, Phys. Rev. B **49**, R1513 (1994).
[39] M. R. Bhat, Nucl. Data Sheets **88**, 417 (1999).
[40] E. N. Shipley, R. E. Holland, and F. J. Lynch, Phys. Rev. **182**, 1165 (1969).
[41] B. Lengeler, C. Schroer, J. Tümmler, B. Benner, M. Richwin, A. Snigirev, I. Snigireva, and M. Drakopoulos, J. Synchrotron Radiat. **6**, 1153 (1999).