

Nuclear Resonant Scattering from ^{161}Dy at 25.65 keV

Nuclear resonant scattering (NRS) of synchrotron radiation [1] provides several methods for investigating materials. On one hand, time differential measurements allow access to nearly all of the hyperfine and materials science information of conventional Mössbauer experiments, with added structural information possible due to the use of a scattering geometry and the extremely brilliant X-ray beam. On the other, nuclear inelastic scattering (NIS) uses the narrow nuclear resonance as a probe of the nuclear motions within a material, allowing access to phonon spectra on meV energy scales. Most NRS experiments provide information specifically about the excited resonant nuclei (*i.e.*, hyperfine fields at the resonant nucleus, phonons that include the resonant nucleus, structural and motional correlations among the resonant nuclei). This specificity allows one to obtain very precise information. It also provides clear impetus for extending the number of available transitions beyond the few that are now commonly used.

NRS experiments with new resonant transitions are largely limited by instrumentation. While

stronger synchrotron sources always present new opportunities, in fact present sources are strong enough (at least for low to mid-range X-ray energies) to provide rather high flux in the resonant bandwidth of many transitions. Using that flux, without large losses either in optics or detection, however, is a significant challenge. Here we describe a new setup, commissioned at **BL35XU**, for nuclear scattering with the 25.65 keV resonance of ^{161}Dy . In principle, this resonance is convenient for synchrotron based studies, having a relatively long (42 ns) lifetime and a large cross-section (low internal conversion). In addition, the difficulties in obtaining narrow lines in conventional (radioactive source) Mössbauer measurements make the synchrotron based work especially appealing. However, practically, significant instrumentation work is required to make experiments feasible. We have developed a new monochromator and a new detector optimized for this resonance [2].

Our monochromator uses a thin coupling crystal placed inside of a high order channel cut crystal operating near backscattering (see Fig. 1). By using the coupling crystal both in reflection and transmission, this design allows one to get extremely close to backscattering (Bragg angles near 90 degrees) without the very big crystals that would be needed for previous designs [3]. Thus,

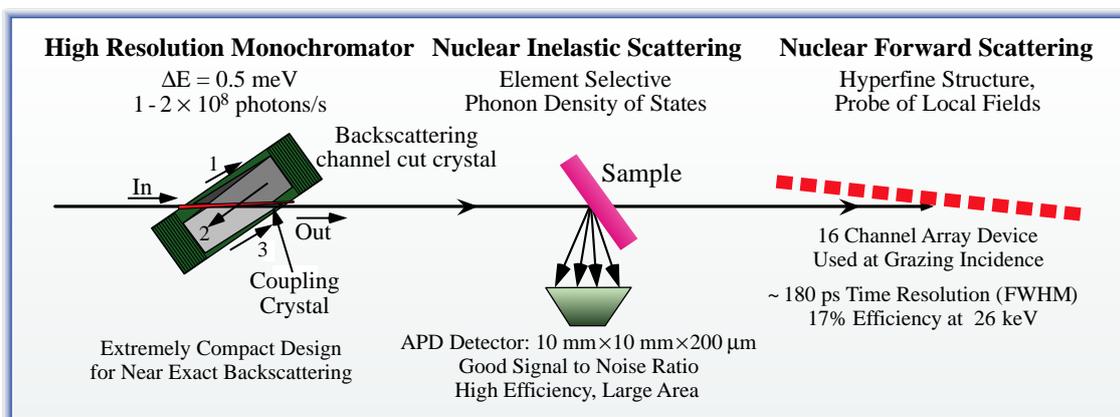


Fig. 1. Schematic of the setup for nuclear resonant scattering with ^{161}Dy summarizing properties of important components.

one can take full advantage of the large angular acceptances available near backscattering, while retaining a compact, in-line, geometry. In particular, working at 25.65 keV (using the (6 2 0) and (18 12 6) reflections – Bragg angles of 16.3 and 87.4 degrees, respectively) we have obtained a 0.52 meV bandwidth ($\Delta E/E = 2 \times 10^{-8}$) and a peak throughput of about 2×10^8 photons/s, both in good agreement with theory (additional discussion can be found in [4]). The monochromator was used to measure nuclear inelastic scattering from several samples, and results from a DyB_2C_2 sample (natural abundance, 19% ^{161}Dy) are shown in Fig. 2. In particular, the relatively simple phonon density of states makes it easy to identify the various multi-phonon contributions appearing at high temperature, while the inset shows the derived density of states, with some softening evidence at room temperature.

In general, this monochromator design should be easy to use in the 20 - 30 keV range and work on a monochromator for the Sn resonance at 24 keV is in progress [5]. At lower energies, absorption

in the silicon coupling crystal becomes a problem, but this might be avoided by using a less absorbing material (e.g. diamond or beryllium). Extension to higher energy in silicon is also possible, but, the backscattering reflectivity of the silicon falls off quickly, so other materials (e.g. sapphire) with a higher Debye temperature might be advantageous, if a suitable quality crystals can be found. At exact backscattering, this design becomes reminiscent of an X-ray Fabry-Perot interferometer, suggesting a way of controlling the coupling into/out of such an interferometer that is independent of the backscattering mirrors.

Development of a proper detector for nuclear forward scattering (NFS) from ^{161}Dy is challenging. This is because one needs both high efficiency (as the resonance is narrow and count rates are small) and extremely good time resolution (as the hyperfine splitting of the ^{161}Dy resonance can lead to beat frequencies of ~ 10 GHz). These two conditions are usually mutually exclusive in silicon avalanche photodiodes (APDs) since making them thicker to improve the efficiency degrades the time resolution:

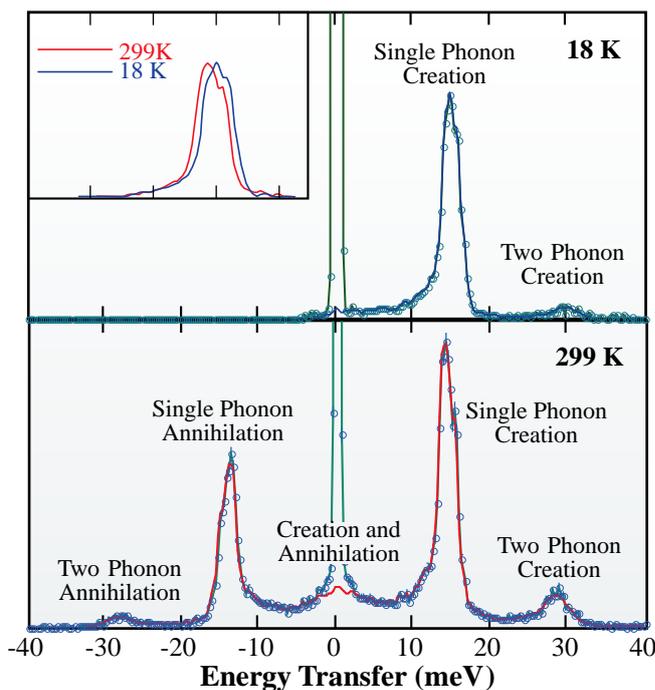


Fig. 2. Nuclear inelastic scattering from DyB_2C_2 at low temperature and room temperature. The solid lines in the figure are calculations based on the derived partial density of states DOS (shown in the inset – 22.5 meV full scale). See text for discussion.

the ratio of the active thickness to the time resolution for an APDs is approximately constant, and equal to the saturation drift velocity of the electrons in the APD ($\sim 100 \mu\text{m}/\text{ns}$ for silicon). However, this trade-off may be circumvented using an array of thin devices at grazing incidence, allowing a long path length through the silicon, without making the electron transit time spread longer. An array of 16 elements (each $1 \times 2.5 \text{ mm}^2$ on a 1.1 mm pitch) allowed us to achieve 180 ps resolution with about

a 0.5 mm path length in the silicon, corresponding to $\sim 17\%$ efficiency at 25.65 keV. While the 180 ps resolution is not quite sufficient to resolve the fastest beats from ^{161}Dy it is sufficient so that there are no isolated lines in the response – beats will appear from all excited levels. The time response measured from a Dy foil at low temperature is shown in Fig. 3, the excellent time resolution is clearly evident, and the general quality of the data is confirmed by the good agreement with theory.

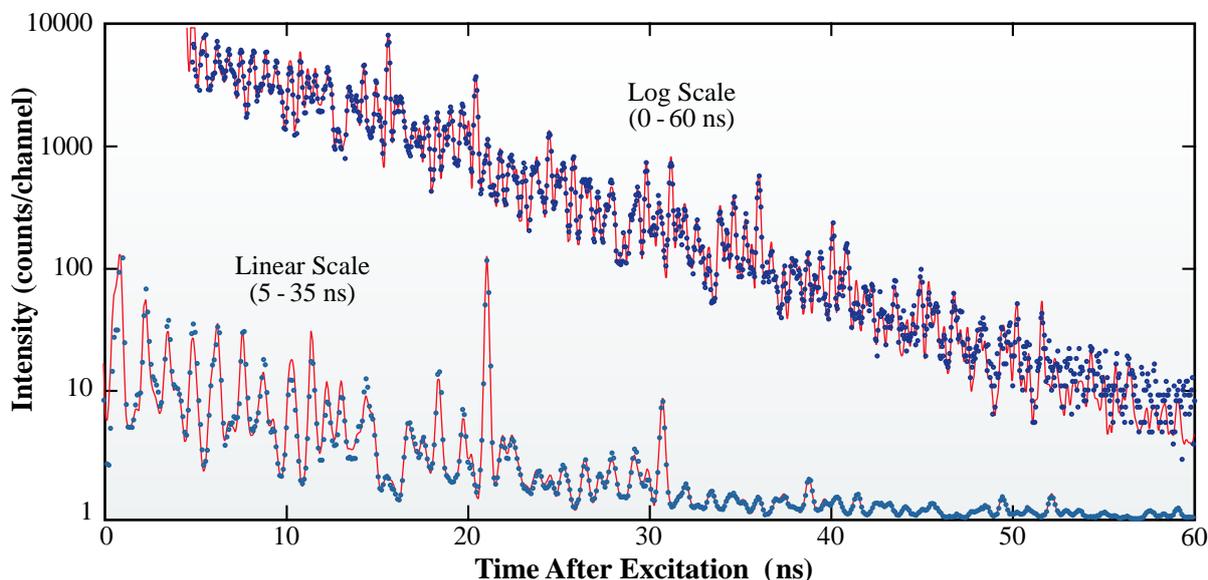


Fig. 3. Nuclear forward scattering from a (non-enriched) ^{161}Dy foil at low temperature. The agreement between the fit (solid line) and the data serve to highlight the good performance of the detector. Note both log and linear plots are shown and that the axis labels are for the log plot.

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