



	Experiment title: Enhancement of the Dynamic Range of Synchrotron Radiation Based Perturbed Angular Correlation	Experiment number: MI-639
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Report:

Beside the established methods of Nuclear Forward Scattering (NFS) and Nuclear Inelastic Scattering (NIS), a new method based on spatially incoherent, single-nucleus scattering is presently under development, Synchrotron Radiation based Perturbed Angular Correlation (SRPAC). SRPAC has been recently applied in in-house research to investigate glass dynamics under conditions of slow to medium reorientational relaxation [1]. The aim of the present experiment was to perform systematic SRPAC measurements in a wider temperature range in order to test the method in a larger dynamic range.

For this purpose, the experimental setup (compare Fig.1) was modified in three important points:

- In order to increase the countrate, a larger APD detector (area $10 \times 10 \text{ mm}^2$) was placed in 90° scattering geometry at closer distance below the sample, yielding a gain in intensity by a factor of ~ 36 . A slight decrease of the contrast due to angular average had to be tolerated.
- In order to reduce the load of the prompt radiation on the detector, the bandwidth of the incident beam was reduced to $\sim 6 \text{ meV}$ by using a standard high-resolution monochromator.
- In order to increase the sensitivity, the experiment was performed in the single bunch mode, allowing for a large observation window. This permits to push both limits of the dynamic range: the lower limit of slow relaxation, where a very weak damping of the quantum beat modulation has to be observed, and the upper limit of fast relaxation, where a very slow approach of the decay to the natural decay has to be followed. Both effects are better seen over a large time window.

In the experiment the dynamics of a model substance, the molecular glass former dibutyl phthalate (caloric glass transition at $\sim 179 \text{ K}$), was monitored via nuclear scattering on inserted ferrocene probes enriched in ^{57}Fe . The time evolutions of SRPAC and NFS were measured in parallel. SRPAC data were taken in the temperature range $160\text{-}330 \text{ K}$, i.e. from the glassy to the liquid state, and NFS data up to 210 K , where the Lamb-Mössbauer factor vanishes. The incident radiation was centered at nuclear resonance. Integral SRPAC countrates were typically $30/\text{s}$. Because of the exponential decay of the SRPAC time evolutions with the ^{57}Fe natural life time $\tau_0=141 \text{ ns}$, data collection was limited to a time window of 800 ns .

Fig.2 shows typical time evolutions. In the upper part, the time evolutions of SRPAC and NFS are compared at 206 K . The SRPAC data exhibit a natural decay, whereas the decay of the NFS data is accelerated due to collective scattering and relaxation. Both time evolutions are modulated by a quantum beat with single frequency Ω corresponding to the quadrupole splitting of ^{57}Fe in ferrocene. The modulations are in antiphase due to different scattering geometries, in plane for NFS and 90° out of plane for SRPAC. In the lower part of

Fig.2, time evolutions of the SRPAC anisotropy $2A_{22}G_{22}(t)$, defined by $I(t) \propto \exp(-t/\tau_0)(1-2A_{22}G_{22}(t))$, are shown for different temperatures. Two relaxation regimes can be distinguished:

- In the slow relaxation limit (< 230 K), a pronounced quantum beat is discernible. At 180 K the beat exhibits only a very weak damping. There is, however, a typical disturbance at early times which results from 90° Rayleigh scattering of the NFS channel which is still intense at this temperature. For rising temperatures, the damping of the quantum beat becomes much stronger, which can be described by increasing rotational relaxation rates λ_r . In spite of the fading of the signal, beat period and damping can still be distinguished in this regime, and Ω and λ_r can be determined independently.
- In the fast relaxation limit (> 250 K), the quantum beat is not discernible any more, only the slow approach of the decay towards the natural exponential decay can be observed (Abragam-Pound limit). In this regime, Ω and λ_r cannot be determined independently, and the evaluation of λ_r depends on assumptions on Ω .

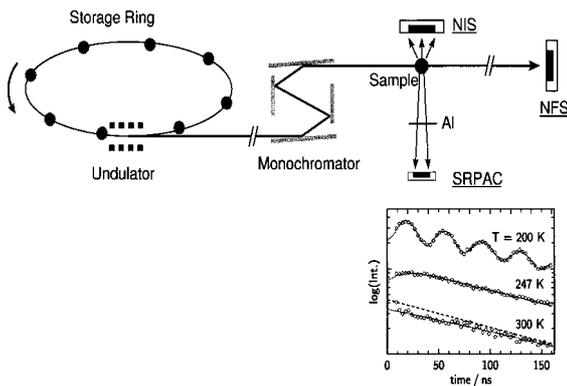


Fig.1 Experimental setup

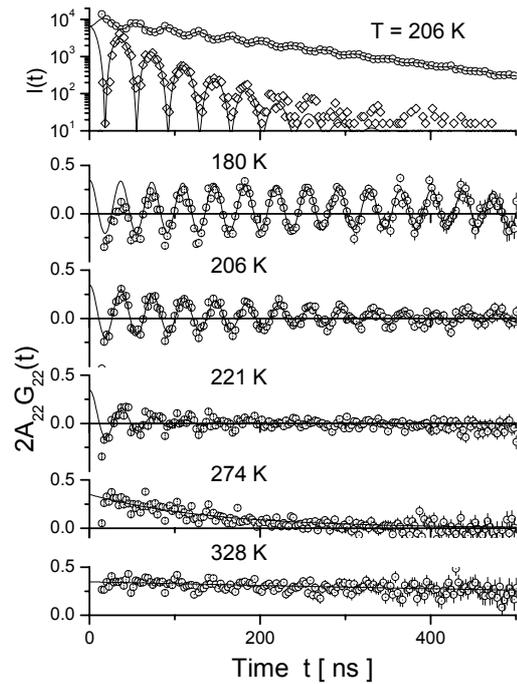


Fig.2 Time evolutions for NFS and SRPAC

From the SRPAC data, the temperature dependence of the pure rotational relaxation rate λ_r was extracted. It extended from a lower limit of $\sim 2 \cdot 10^6$ Hz (relaxation time 500 ns) at 160-180 K to an upper limit of $4 \cdot 10^{10}$ Hz (relaxation time 25 ps) at 330 K. Thus altogether a dynamic range of $2 \cdot 10^4$ was covered in the experiment, largely corresponding to our expectations. Clearly these limits can still be pushed somewhat further by counting longer times in order to reach better statistics for the cases of extremely slow or fast relaxation.

In parallel, the total relaxation rates $\lambda = \lambda_r + \lambda_t$ were determined from the NFS data which are sensitive to both rotational and translational relaxation on an atomic length scale. A comparison of the SRPAC and NFS relaxation rates allowed us to extract the temperature dependence of the translational relaxation rates λ_t . This way it became possible to analyze the rotational and translational dynamics of the ferrocene probes separately and to use these results to interpret the dynamics of the host dibutyl phthalate known from dielectric spectroscopy. A detailed evaluation of the data and an interpretation of the results in the context of glass physics is under way [2,3].

[1] I. Sergueev et al., ESRF Highlights 2002, p.61

[2] I. Sergueev, PhD-Thesis, TUM 2003, in preparation

[3] I. Sergueev et al., to be published