ESRF	**Experiment title:** Quasielastic Scattering by Time Domain Interferometry: Comparison of the Quantum Beat and Radiative Coupling Regimes	**Experiment number**: MI-533
Beamline: ID22-N	**Date of experiment**: from: 25-10-01 to: 30-10-01	**Date of report**: 28.08.02
Shifts: 15	**Local contact(s)**: Dr.E.Schreier, Dr.O.Leupold	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dr. U. van Bürck*, Techn.Univ.München, Physik E13, D-85748 Garching, Germany

Prof. Dr. G.V. Smirnov, RRC Kurchatov-Institute, Moscow 123182, Russia

Dr. H. Franz*, HASYLAB at DESY, D-22603 Hamburg, Germany

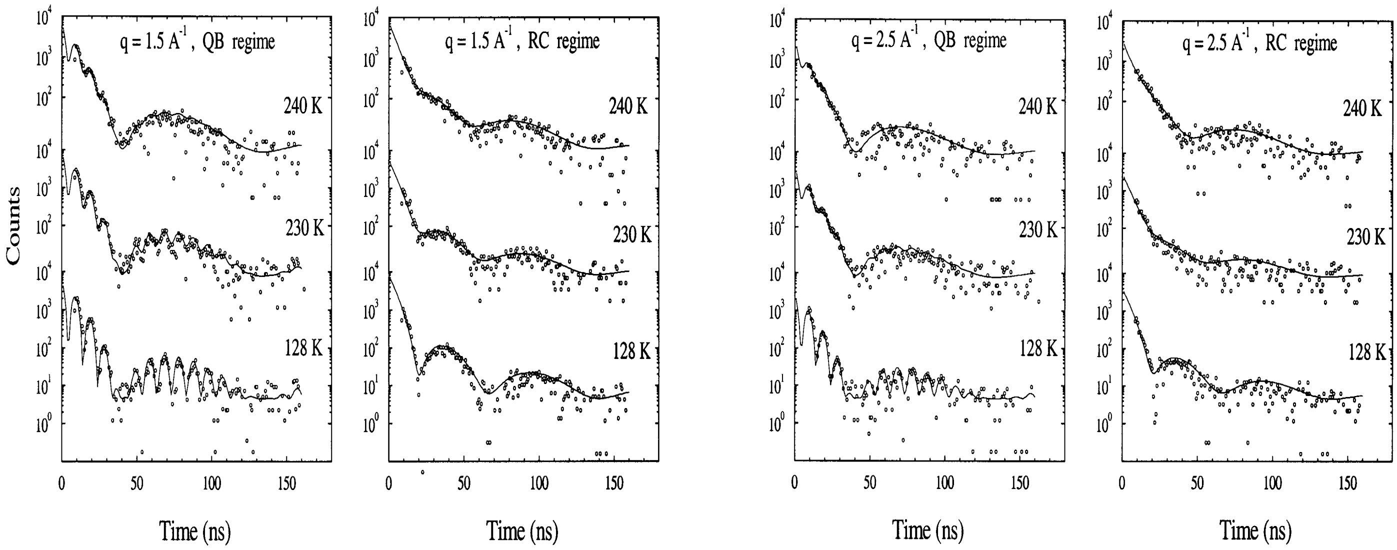
Dr. T. Asthalter*, Universität Stuttgart, Inst.Phys.Chem., D-70569 Stuttgart, Germany

Report:

The extreme sharpness of nuclear resonant scattering is a challenge calling for applications e.g. in the field of slow atomic motions. A method not limited to nuclear resonant samples is time domain interferometry (TDI), where the quasielastic scattering of synchrotron radiation (SR) by a non-resonant sample is analyzed by means of the delayed nuclear forward scattering (NFS) from two identical resonant targets. One target is mounted upstream and the other downstream of the sample. In case of a stiff sample, when one nuclear target is at rest and the other moved at large constant velocity, interference of the waves scattered by the two targets leads to a fast quantum beat (QB) which modulates the slow dynamical beat (DB) of the single-target response. When both nuclear targets are at rest, the wave scattered by the upstream target can be scattered again by the downstream target, and this so-called radiative coupling (RC) creates a DB corresponding to twice the optical thickness (double-target DB). If however the sample exhibits dynamics, the phase of the radiation scattered by the upstream target becomes perturbed by the quasielastic scattering in the sample. This perturbation destroys interference and radiation coupling, leading for TDI in the QB-regime to a fading of the QB [1], whereas for TDI in the RC-regime a transition from the double-target DB to a single-target DB is expected [2]. These two regimes of TDI were to be compared in the present experiment.

The experimental set-up [1] was identical for both methods. The incident SR was monochromatized to ~6meV by a high resolution monochromator Si(422)/Si(1222) in nested geometry. Special care had been taken to minimize vibrations from the vacuum pump and from the constant velocity Mössbauer drive possibly affecting the triple system of the two targets and the scattering sample mounted in a N_2 bath cryostat. Also the radiation background reaching the detectors for the scattered beams was carefully minimized. In the experiment, the quasielastic scattering from a sample of glycerol at temperatures T ~ 128, 230 and 240 K was analyzed by TDI both in the QB-regime and the RC-regime, using as targets a set of two 6 µm thick stainless steel foils enriched to 95% in ^{57}Fe. Measurements at different scattering angles ~ 8, 12, 16 and 20 degrees corresponding to scattering vectors of magnitudes q ~ 1, 1.5, 2 and 2.5 $Å^{-1}$ were performed in parallel using a segmented avalanche photodiode (APD) area detector [3]. The motion of the constant velocity drive was all time monitored via NFS by the target on the drive and another ~ 6 µm stainless steel target rigidly mounted in forward direction. The constant velocity was set to zero in the RC-regime, and to ~ ±9 mm/s in the QB-regime.

The experiment suffered from unexpectedly low delayed countrates ≤ 1 /s in the scattering detector, resulting from an unidentified ~ 50% lack of intensity of the beamline ID22-N and from jump instabilities probably occuring in our detector electronics. We estimate that in stable conditions and at more powerful beamlines like e.g. ID18 TDI experiments could be performed with delayed countrates in the order of at least ~ 10/s. Such countrates would allow to use TDI almost as a standard method.



In spite of the uncomfortably low intensity of the present experiment, taking data for ~9h in each regime and for each temperature allowed to demonstrate TDI in the RC-regime for the first time and to compare TDI in the RC- and the QB-regimes. As an example, we show in the figure the time evolutions measured at q ~ 1.5 Å^{-1} (left block) and ~ 2.5 Å^{-1} (right block) for the QB- and RC-regimes at different temperatures, with background substracted. At 128 K, where the scattering sample is stiff, the time evolutions in both regimes correspond to the instrumental functions measured in forward direction (compare Figs. 2h and 2a of [4]). At higher temperatures the sample exhibits dynamics, which gradually destroys interference and radiative coupling. In the QB-regime, a fading of the QB [1] is observed, which varies both with temperature and scattering angle. In the RC-regime, a characteristic transformation from a double-target DB (see e.g. 1.5 Å^{-1} /128 K: first DB minimum at ~ 20 ns) to almost a single-target DB (see 2.5 Å^{-1} /240 K: first DB minimum at ~ 40 ns) can be observed in parallel.

This qualitative comparison has been substantiated by a preliminary computer analysis. The solid lines in the figure are simulations based on Eqs. (32,33) of [2] in the approximation of weak scattering, where a Kohlrausch type of relaxation was assumed with parameter β = 0.7 [5] fixed. At the given poor accuracy of the data, all time evolutions could be well described with a dominating self-part of the van Hove pair-correlation function. The most important point is that it was possible to find parameters by which the two sets of completely different time evolutions for the QB- and the RC-regimes were consistently fitted.

As for the sensitivity of TDI in the two different regimes, the following can be said. Since the QB is a very pronounced feature of the time evolution measured in the QB-regime, its fading due to sample dynamics is more easily recognized during the experiment than the corresponding changes of the DB in the RC-regime. The computer analysis, by contrast, is more sensitive to relaxation parameters when the time evolutions were measured in the RC-regime. Therefore in future at least some first measurements should be performed in the QB-regime for on-line orientation, and then the majority of the measurements in the RC regime. But we also feel that the consistent fit of such two different data sets increases the reliability of the data evaluation.

[1] A.Q.R. Baron *et al.*, Phys.Rev.Lett. 79 (1997) 2823.

[2] G.V. Smirnov, V.G. Kohn, and W. Petry, Phys.Rev. B 63 (2001) 144303.

[3] H. Franz *et al.*, HASYLAB Ann.Report 2001, p.101.

[4] U. van Bürck *et al.*, Phys.Rev. A 61 (2000) 013803.

[5] J. Wuttke, W. Petry, and S. Pouget, J.Chem.Phys. 105 (1996) 5177.