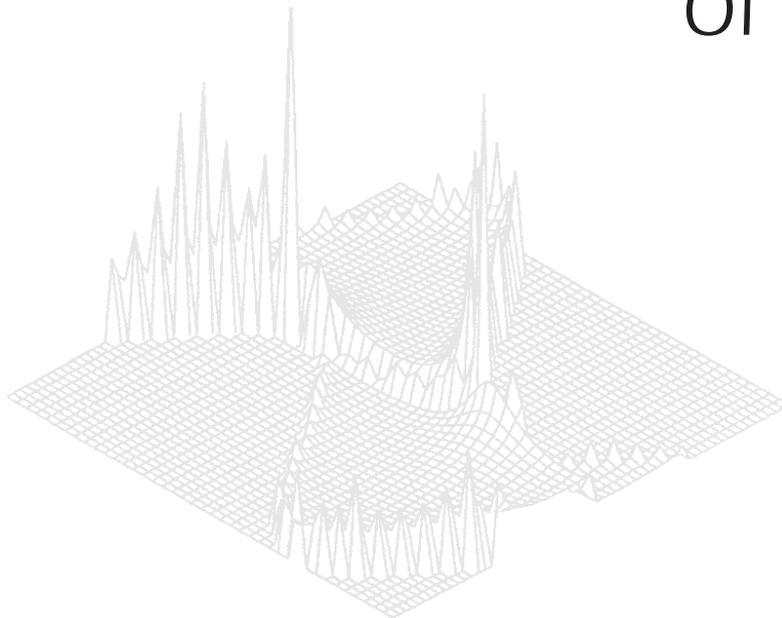

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Spatial Distribution of Magnetic and Nonmagnetic Phases in Nuclear Forward Scattering of Synchrotron Radiation*

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Abstract

Nuclear forward scattering of synchrotron radiation has been demonstrated to permit conclusions on the spatial distribution of scattering centres. An outline of the method is given and implications for samples exhibiting domain structure are discussed.

1. Introduction

It was Ruby (1974) who first proposed using synchrotron radiation (SR) to excite nuclear levels. The central idea was to combine the advantages of SR with the high energy resolution of the Mössbauer effect. After the first successful experiments (Gerda *et al.* 1985, 1986; Rüffer *et al.* 1987), new monochromatisation techniques were introduced (Faigel *et al.* 1987). This opened the way to pass from a complicated crystal reflection geometry to a simple transmission geometry which permits the study of polycrystalline materials. Nuclear forward scattering (NFS) of SR was first demonstrated by Hastings *et al.* (1991) and van Bürck *et al.* (1992).

2. Basic Features of NFS

The scattering of SR by the electron shell of the sample atoms will be almost instantaneous. Nuclear scattering, on the other hand, will be delayed, a characteristic delay time being the mean lifetime of the nuclear excited states involved in the scattering process. The pulsed time structure of SR thus opens the possibility to separate electronic and nuclear scattering and to study the decay of excited nuclear levels. A flash of synchrotron light creates a collective nuclear excited state. The radiation pulse that excites the nuclei is typically short compared to the natural lifetime of the individual nuclear levels. The frequency bandwidth of the radiation pulse, on the other hand, is by several

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orders of magnitude broader than the natural linewidth of the nuclear decay. In general all allowed nuclear transitions are therefore excited simultaneously. The intensity of the successive nuclear decay is then modulated by ‘quantum beats’. The frequency of such beats thereby reflects the hyperfine splitting of the nuclei in the sample and can serve as a quantitative measure of the splitting. In a thick sample the decay of a collective excited state proceeds much faster than the natural decay (speed-up). The time evolution of the forward scattered intensity is additionally modulated by a beat structure (‘dynamical beating’) which depends on the number of resonantly excited nuclei in the crystal. These two effects, the speed-up of the nuclear decay and the dynamical beating, are multiscattering effects. For details on the theory of NFS see e.g. Kagan *et al.* (1979) and Smirnov (1996).

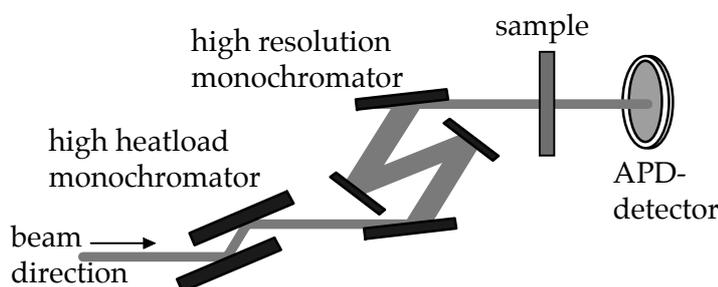


Fig. 1. Schematic of a typical set-up for nuclear forward scattering experiments. The incoming beam is reduced in bandwidth from ≈ 300 eV to several meV in two steps, scattered by the sample and detected by a fast avalanche photodiode detector (APD).

3. Experimental Conditions

The advent of third generation SR sources has given new impetus to the field. A new undulator station (ID 18) dedicated to the exploration of nuclear resonance techniques came into operation in March 1995 at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France (Rüffer and Chumakov 1996). Undulators are sources of high brilliance, i.e. they provide high photon flux in a given bandwidth with small beam size and small beam divergence. The bandwidth of the ID 18 undulator, which is designed for the investigation of the 14.413 keV resonance of ^{57}Fe , is ≈ 300 eV, while only a bandwidth of $\approx 0.5 \mu\text{eV}$ can be used to excite the hyperfine split levels. This poses insurmountable problems to almost any known detector system. The radiation is therefore monochromatised in two steps (see Fig. 1). A water-cooled Si(111) double crystal monochromator is used to reduce the bandwidth to ≈ 2 eV. In a second step the bandwidth is further reduced down to several meV by crystal monochromators using high indexed reflections and asymmetric cut crystals. Fig. 1 shows the ‘nested design’ (Ishikawa *et al.* 1992; Toellner *et al.* 1992) where two channel cut crystals are employed. The outer two reflecting surfaces are asymmetrically cut in order to match the divergence of the synchrotron beam to the acceptance of the inner high-indexed reflections. The outgoing beam is scattered by the sample and detected by a fast avalanche photodiode detector (Baron 1995).

4. Coherence Properties

The possibility of observing quantum beats or dynamical beating in NFS is governed by the coherence properties of the radiation. These properties are of interest for samples having different nuclear environments as they occur for example in different crystallographic phases or magnetic domains. Scattering from nuclei that lie on a direct line from source to detector will always add coherently, i.e. the observed intensity will be a squared sum of scattering amplitudes. For nuclei that are separated perpendicular to the beam direction, incoherent addition of scattering will also be observed where the scattering centres are separated by more than the transverse coherence length. The transverse coherence length is a function of the scattering geometry, as has been shown for NFS recently (Baron *et al.* 1996).

5. An Experimental Example

The impact of the specific experimental geometry on the coherence properties of the scattered radiation observed in an NFS experiment first showed up during a NFS investigation of the $\alpha \rightarrow \epsilon$ transition in iron (Grünsteudel *et al.* 1996). The pressure driven phase transformation from magnetic α -iron to nonmagnetic ϵ -iron at room temperature was first reported by Bancroft *et al.* (1956). The appearance of ϵ -iron in the sample above a pressure of 13 GPa is directly evidenced in energy domain Mössbauer spectra as a single line in the centre of the six line spectrum of α -Fe (Pipkorn *et al.* 1964; Williamson *et al.* 1972; Taylor *et al.* 1991). With increasing pressure the center line becomes more and more intense until at pressures higher than about 25 GPa only the ϵ -phase remains. Different pressure values exist in the literature for both onset and completion of the transition. A systematic dependence of these pressure values on the shear strength of the pressure transmitting media employed in different studies has been reported (von Bargen and Boehler 1990). The $\alpha \rightarrow \epsilon$ transition is an example where regions with different nuclear environments in the sample are created through a crystallographic phase transition and are easily distinguishable by their magnetic behaviour.

The time evolution of NFS by an ^{57}Fe foil at 15 GPa is shown in Fig. 2. The experimental conditions are those described in Grünsteudel *et al.* (1996). At this pressure both α - and ϵ -phases coexist in the sample. The up and down of the intensity evolution with time are quantum beats which evidence the magnetic hyperfine splitting in α -iron. The beat contrast is much less pronounced than could be expected for α -iron, which is due to the presence of the ϵ -iron in the sample. We find that, as discussed above, scattering from α - and ϵ -phases contributes to the measured time response through both coherent and incoherent addition. The contributions are illustrated in Fig. 3. Calculated time responses are shown for (a) α -iron and (b) ϵ -iron. A coherent addition (sum of amplitudes) of both responses is shown in (c) for 50% α -iron and 50% ϵ -iron. Note the drastic change in the beat structure and the decrease in the quantum beat contrast compared to the time response of α -Fe. This curve simulates scattering from sample regions that either lie ‘behind each other’ when seen with the beam or that are separated transversely to the beam not more than the transverse coherence length. Such a time response can be expected for a sample where

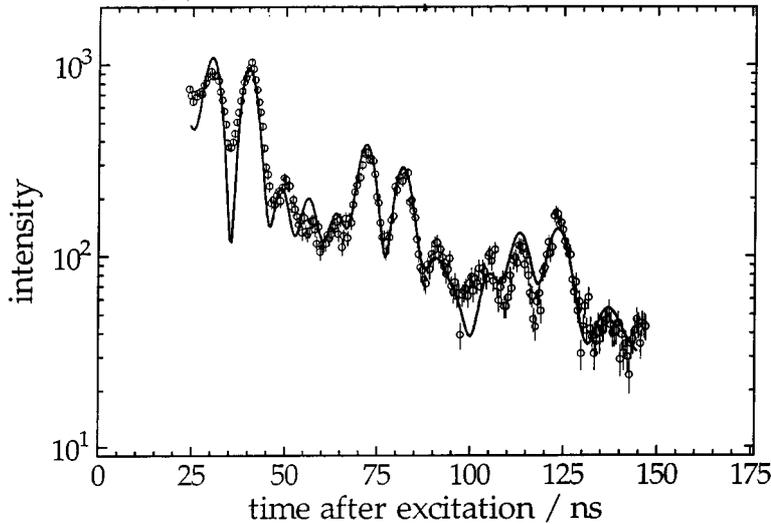


Fig. 2. Time evolution of nuclear forward scattering of synchrotron radiation measured with a $3\ \mu\text{m}$ ^{57}Fe foil at 15 GPa. The curve is an incoherent sum using the coherent time responses of α -Fe (21%), ϵ -Fe (38%) and the *coherent* sum of both (41%).

regions with different nuclear environments are ‘well mixed’. Part (d) shows an incoherent addition (sum of intensities) for the same α/ϵ ratio as in (c). In such a sample different regions would be separated more than the transverse coherence length and cannot lie ‘behind each other’. Here only the high–low contrast of the quantum beats is reduced while the beat frequencies remain the same.

The curve through the data in Fig. 2 is a least squares fit using a weighted sum of three contributions: Scattering from α -Fe (analog to Fig. 3a) contributes with 21% and from ϵ -Fe (analog to Fig. 3b) with 38%. The remaining 41% of the intensity is due to scattering from regions in the sample where scattering from both phases adds coherently (analog Fig. 3c considering 37% α -Fe and 63% ϵ -Fe). The best adjustment was obtained assuming a texture of the magnetisation of the α -phase. All calculations of coherent time responses shown in Figs 2 and 3 have been performed using the Conuss program (Sturhahn and Gerdau 1994).

6. Discussion

The key to an interpretation of these numbers is the effective transverse coherence length, which in this experiment was of the order of $\approx 10\ \text{\AA}$ (calculated after Baron *et al.* 1996). In other words, scattering from nuclei separated transversely by more than $\approx 10\ \text{\AA}$ should not add coherently. The fit to the data, on the other hand, shows a significantly large contribution of coherent scattering (41%). Since the effective transverse coherence length is by several orders of magnitude smaller than the size of the sample perpendicular to the beam ($\approx 100\ \mu\text{m}$) we can clearly state that the α - and ϵ -phases coexist in the sample and lie ‘behind each other’. This implies that regions of either phase cannot be larger than about $1\ \mu\text{m}$. This kind of information cannot be obtained with conventional Mössbauer spectroscopy. From the various contributions we derive a total amount of 36% and 64% of α -phase and ϵ -phase, respectively, in

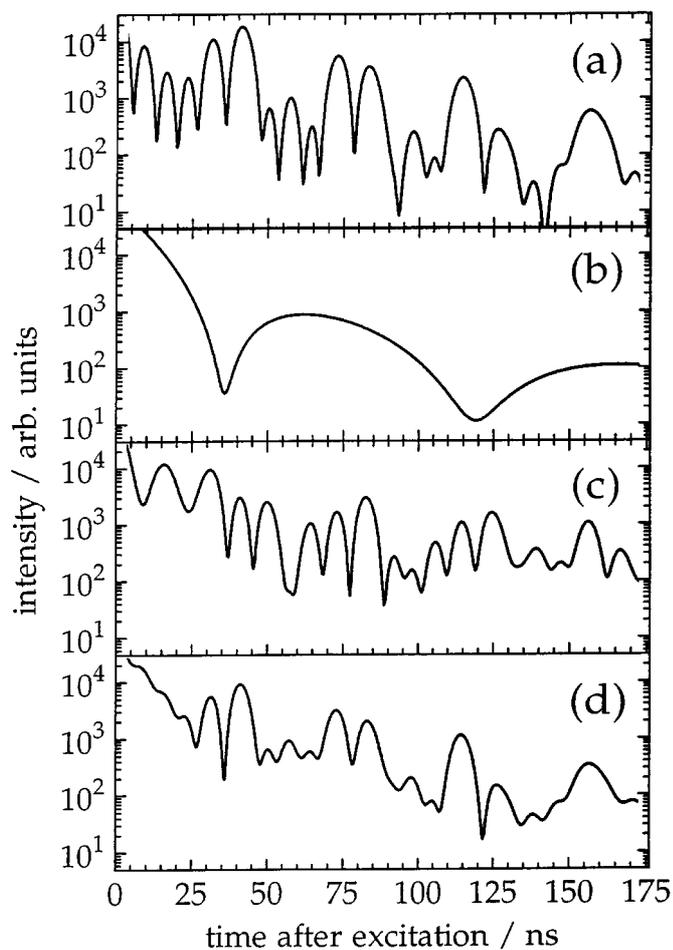


Fig. 3. Calculated time evolution of nuclear forward scattering of synchrotron radiation for (a) α -Fe and (b) ϵ -Fe. Also shown are (c) the coherent and (d) the incoherent sum of both time responses for 50% α -Fe and 50% ϵ -Fe.

agreement with results from conventional Mössbauer spectroscopy at the same sample (Grünsteudel *et al.* 1996). The experiments on the $\alpha \rightarrow \epsilon$ transition in iron have been further improved (Grünsteudel 1997) and have confirmed the evaluation procedure described above.

7. Conclusion

The superior brilliance of SR compared to radioactive sources makes NFS a powerful tool for high pressure investigations probing magnetism. As shown by the example presented here it is worth while to further investigate NFS as a novel method of probing the spatial distribution of varying nuclear environments in bulk material. The investigations on the transverse coherence in NFS (Baron *et al.* 1996) imply that the transverse coherence length can be chosen for the

experiment to a certain extent. With an appropriate choice of slit systems the transverse coherence length can be varied and serve as a probe of domain structure. In combination with the recent development of nuclear small angle scattering (Shvyd'ko *et al.* 1996) new information on such systems may be obtained.

Acknowledgments

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