NEUTRON DIFFRACTION UNDER VERY HIGH PRESSURES: EXPERIMENTS IN THE 40 GPa PRESSURE RANGE.

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The role of high pressures in solid state physics is essentially twofold: 1) pressure varies interatomic distances in a controlled way and therefore could be used to verify theoretical models, 2) it induces new phases, which may exhibit unusual characteristics. In practice, the possibility of using pressure strongly depends on the range of pressure needed and on the technical tools required to reach it. In soft matter, even pressures of less than 1 GPa may strongly change the interatomic distances and the physical properties. In contrast, in most solids, much higher pressures (10 GPa or more) are needed to produce significant change of the sample volume ($\Delta V/V > 10\%$). In the recent years, crystal structures studies in the 100 GPa pressure range have strongly benefited from the development of third-generation synchrotron X-ray sources, whereas magnetic studies in the same pressure range are still much less developed. Anomalous magnetic X-ray scattering cannot be used because of the huge absorption of the pressure device, and Mössbauer studies do not provide any information about the long range magnetic order. Neutron diffraction is usually limited to much lower pressures (1–3 GPa). This is because pressures above 10 GPa could be reached only by a drastic decrease of the available sample volume (down to 10^{-4} to 10^{-6} mm^3 in the 100 GPa pressure range). Such a sample volume is well below that usually needed in neutron studies (typically a few mm^3 to a few cm^3).

At the beginning of the 1990's, a project was initiated at the LLB to develop magnetic studies under very high pressure using high intensity neutron diffraction [1]. The steady state reactor with a cold source provides the most convenient q-range for such studies.

The project encompasses both single crystal and powder diffraction under pressure. Single-crystal diffraction yields very detailed information about spin arrangements. Numerous studies using single crystal diffraction under pressures were performed at the LLB during the last years [1,2], most of them on the lifting detector spectrometer 6T2. Very recently a new superconducting magnet and a dilution refrigerator, compatible with the high pressure setup, have become available. Implementing high magnetic fields (up to 8 T) and very low temperatures (down to 30 mK) together with pressures up to 10 GPa opens new possibilities to single crystal studies.

On the other hand, powder diffraction is the most straightforward way to extend neutron studies well above 10 GPa. In this pressure range, single crystals could be easily destroyed by pressure inhomogeneities or by a first order structural transition.

The development of a specialized powder diffractometer, allowing neutron studies to be made at pressures up to 50 GPa, i. e. at higher pressures than in any other neutron facility, was the most ambitious part of the project. During the last three years the G6.1 diffractometer (equipped with a 400-cell multidetector) was transformed in a specialized spectrometer, optimized for the magnetic diffraction of small samples (less than 1 mm^3) under very high pressures. In this high pressure version, G6.1 is equipped with a double–stage focusing system inserted between the monochromator and the sample position (fig 1).

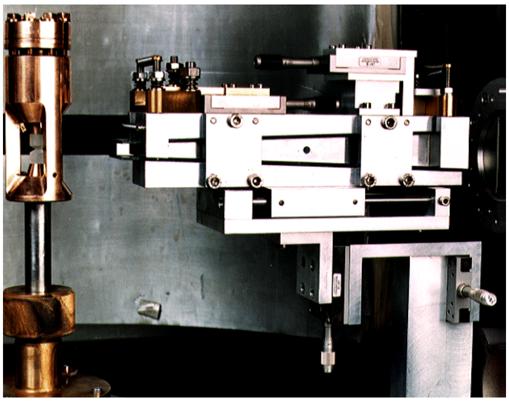

Figure 1. The pressure cell and the focusing device used in the high pressure version of the G61 spectrometer.

The focussing system, consisting of Ni-Ti supermirrors "compresses" the neutron beam both in horizontal and in vertical planes [3]. Using a double-stage system allows us to simulate a curved profile of the mirrors' surface. This is the optimal profile to get both the highest gain in intensity for a given length of the system (limited by the free space between the monochromator and the sample) and a smooth angular distribution of neutrons in the focused beam. The variable angle of focussing makes it possible to choose the optimal trade-off between intensity and resolution for each experiment. The maximal gain in intensity obtained by focusing is 7.

Together with a high intensity of the incident beam, a very low background is crucial to measure small samples. A sophisticated protection and an especially designed cryostat allow us to reduce the background to a very small value – only a few counts/per cell/per hour.

We use pressure cells with sapphire (P < 10 GPa) and diamond anvils (P > 10 GPa). The implementation of the sapphire anvil cells for neutron diffraction is based on over 10 years of experience gained for such experiments in Kurchatov Institute (Moscow). A number of such cells, adapted to low temperature measurements, were developed and implemented in the neutron experiments (single crystal or powder) at the LLB during the last years. One of the most compact cells with sapphire anvils is shown in fig. 2. This cell can be rotated freely inside the cryostat, allowing to study textures or magnetic domain distributions. The cell is compatible with the dilution refrigerator and the superconductive magnet, making possible to use high pressures, magnetic fields and mK-temperatures in the same experiment.

Figure 2. This small pressure cell can be rotated inside a cryostat

The power of the above technique is illustrated here by the study of magnetic interactions in two closely related systems, the europium monochalcogenides and the gadolinium monopnictides. Both families, having the same crystal structure and the same electronic structure ($4f^7$) for the cations (Eu^{2+} and Gd^{3+}) are usually considered as “model systems” to study magnetic interactions in semiconductors and semi–metals. The neutron diffraction patterns of EuTe and GdAs under pressure are shown in figs. 3 and 4.

The pressure strongly modifies the magnetic properties of EuTe, resulting in a sequence of magnetic transitions at pressures around 10 GPa [4]. As the lattice constant decreases, the ferromagnetic exchange interaction increases very rapidly, leading to a transformation of the initial antiferromagnetic order to a ferromagnetic one, followed by an exponential increase of the Curie point [5].

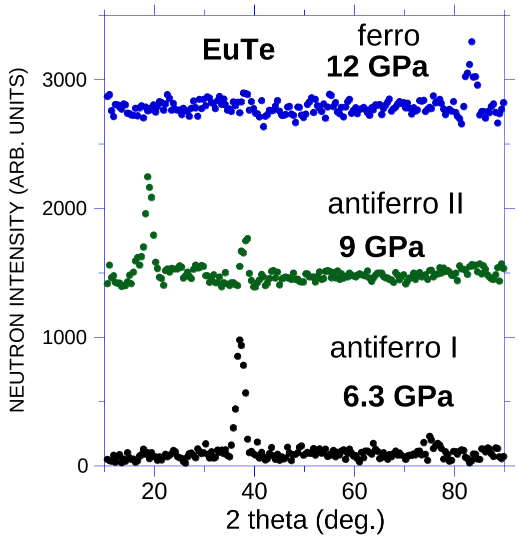

Figure 3. Magnetic neutron diffraction spectra under high pressure in EuTe.

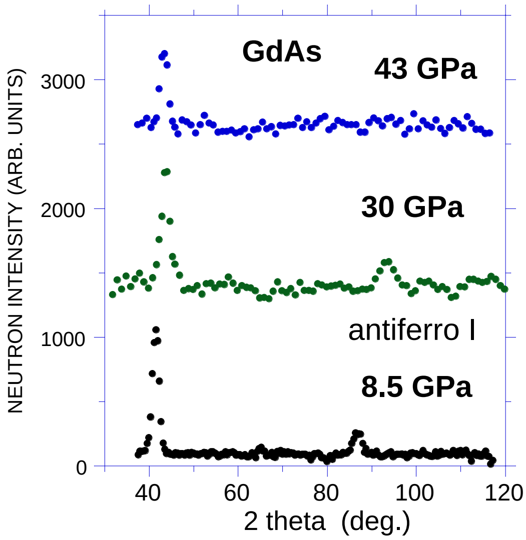

Figure 4. Magnetic neutron diffraction spectra under high pressure in GdAs. The GdAs sample was prepared by A.Ochiai (Univ. of Niigata, Japan).

The GdAs sample, having the same magnetic structure at ambient pressure, shows a very different behavior. Pressure only increases the Néel point. Even at 43 GPa, when the volume decrease reaches about 40%, the compound remains antiferromagnetic. In this measurement, the sample was as small as about 1 mg of weight (0.002 mm^3). This is likely the smallest sample under the highest pressure ever measured with neutrons.

Comparing the two systems (GdAs and EuTe) allows us to see the role of the band structure in the formation of the antiferro- and ferromagnetic exchange between the cations.

In the year 1999, we expect the next upgrade of the high pressure version of the G6.1 spectrometer ("MICRO" diffractometer). A new multidetector, optimized for small samples studies and covering a 7 times larger solid angle, is currently under construction in the European Molecular Biology Laboratory (Grenoble), in collaboration with A. Gabriel. The upgrade of the spectrometer will allow us to increase the available pressure range above 50 GPa and will also provide better possibilities to meet the needs of a growing number of experimental teams, suggesting various studies under pressure in many different fields of physics. Besides the traditional field of magnetism under pressure, new subjects appear, for instance the studies of mesoscopic structures such as the recently discovered carbon nanotubes.

References:

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