High-pressure cells for study of condensed matter by diffraction and inelastic neutron scattering at low temperatures and in strong magnetic fields.

R A Sadykov\textsuperscript{1,2}, Th Strassle\textsuperscript{3}, A Podlesnyak\textsuperscript{3,4}, L Keller\textsuperscript{3}, B Fak\textsuperscript{5,6}, J Mesot\textsuperscript{3}

\textsuperscript{1}Institute for Nuclear Research, Russian Academy of Sciences, Prospekt 60 Letiya Oktiabria 7a, 117312 Moscow, Russia
\textsuperscript{2}Vereschagin Institute for High Pressure Physics, Russian Academy of Sciences, 142190 Troitsk, Moscow, Russia
\textsuperscript{3}Laboratory for Neutron Scattering, ETH&PSI, 5232 Villigen PSI, Switzerland.
\textsuperscript{4}Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
\textsuperscript{5}Commissariat à l’Energie Atomique, Département de Recherche Fondamentale sur la Matière Condensée, SPSMS, 38054 Grenoble, France
\textsuperscript{6}Institut Laue-Langevin 71 avenue des Martyrs 38000 Grenoble, France

E-mail: rsadykov@inr.ru

Abstract. We have developed and implemented series of new original clamp high-pressure cells for neutron diffraction and inelastic neutron scattering at low temperatures. The cells design allows one to place them in the standard cryostats or cryomagnets used on neutron sources. Some results obtained for ZnCr\textsubscript{2}Se\textsubscript{4} are demonstrated as an example.

1. Introduction
The increased interest in quantum critical phenomena, including under pressure caused the need for direct neutron scattering experiments performed at low temperatures in strong magnetic fields and at high pressure. We have made various cylinder-piston high pressure clamp cells (HPCC) for diffraction and inelastic neutron scattering experiments under pressure up to 1.5 GPa at neutron diffractometers DMC (SINQ, PSI, Switzerland) and ILL.

2. Two clamp high pressure cells from zero TiZr alloy
The cells made of nonmagnetic alloys TiZr (zero matrix), as in paper [1], and hard of Al-alloys (V95T, V96T) were successfully used in studies of spiral magnetic structures ZnCr\textsubscript{2}Se\textsubscript{4} as well as magnetic structures in NdAl\textsubscript{3} and Er\textsubscript{0.57}Y\textsubscript{0.43}Co\textsubscript{2} compounds under pressure up to 15 kbar at temperature $T = 4.4$ K [2,3,4]. We present some results obtained in neutron powder diffraction study of the magnetic spiral ZnCr\textsubscript{2}Se\textsubscript{4} structure under high pressure. ZnCr\textsubscript{2}Se\textsubscript{4} undergoes a transition from the paramagnetic state to a simple spiral structure at the Neel temperature $T_N$ of 21K [5,6]. The cube spinel crystal structure has a small tetragonal distortion (approx. $10^{-4}$) at this temperature [7]. This distortion increases with decreasing temperature and the period of the spiral decreases correspondingly [6]. To investigate the pressure dependence of the spiral magnetic structure we have performed a neutron powder diffraction study using two high pressure clamp cells (Figure 1 a,b).

E-mail: rsadykov@inr.ru
Figure 1. Sketches and photos of the two zero-matrix (TiZr alloy) high pressure clamp cells used in PSI-LNS. (a) - small clamp HPCC for use of up to 12 kbar: 1- nut-steel, 2- and 4-support and piston-hard steel, 3- TiZr, zero matrix alloy. (b) - big clamp HPCC for use of up to 15 kbar: 1- support from hard steel, 2- supported inner part -zero alloy TiZr, 3- sample. Diameters of the inner hole are 6 mm for (a)-cell and 1 cm for (b)-cell. Heights of the samples are 25 mm for (a)-cell and 6 mm for (b)-cell.

Figure 2. Neutron diffraction pattern of ZnCr$_2$Se$_4$ at $P = 15$ kbar and $T = 4.4$ K.

Figure 3. The length of the magnetic spiral vs pressure at $T = 4.4$ K in ZnCr$_2$Se$_4$.

Figure 4. The Neel temperature vs pressure in ZnCr$_2$Se$_4$. 
A powder sample of semi-conducting materials was placed into the cell, then the cell was mounted in a hydraulic press. The press generated the pressure required for a particular experiment. This pressure was fixed with the help of clamping nuts. Next, the cell was taken out of the press and put into a cryostat, where it was cooled to a required temperature. Neutron diffraction patterns were obtained on the diffractometer DMC at SINQ. Figure 2 shows a typical neutron diffraction pattern at $T = 4.4$ K and $P = 15$ kbar. The length (period) of the spiral determined from shift 000($\pm$) satellite is plotted in Figure 3 as a function of pressure at 4.4 K. The neutron diffraction study of the simple spiral structure of ZnCr$_2$Se$_4$ mixed with NaCl powder and liquid Fluorinert was made in the temperature and pressure ranges 4.4-50 K and 0-15 kbar. The sample was put in thin lead foil cylinder for decreasing the friction of the piston in channel of the cell. The actual pressure in the sample was determined by comparing the NaCl peak position with the values of its compressibility found in [8]. From the obtained results we can conclude that the period of the spiral structure decreases (Figure 3) and consequently the angle of the spiral magnetic structure increases under pressure up to 15 kbar at $T = 4.4$ K. The Neel temperature increases under pressure up to 36 K at $P = 15$ kbar (Figure 4).

The lattice parameter of ZnCr$_2$Se$_4$ at $T = 4.4$ K changes with increase of the pressure from 10.4712(0.0013) Å at $P = 0$ up to 10.3991(0.0011) Å at $P = 15$k bar. Accordingly, the increase of both $T_N$ and the spirals angle may be understood if the antiferromagnetic superexchange interactions between the Cr$^{3+}$ ions increase more stronger than the ferromagnetic ones with decrease of the sample volume under pressure.

3. Nonmagnetic high-pressure clamp cell from NiCrAl alloy

A new high-pressure cell made of a strong NiCrAl alloy [9] (Figure 5) was used to study a pressure dependence of the spiral magnetic structure of the itinerant electron magnet MnSi. This cell was nonmagnetic down to 2 K and no diffraction pattern from the cell was noticed in magnetic fields up to 6 Tesla. A high sensitivity to deviations from hydrostatic conditions which may be present in the clamping cells is one of the main problems with measuring the magnetic spiral structure of MnSi under pressure. To improve hydrostatic performance, the crystal was placed inside the Teflon cell directly in the 50-50% FC87 / FC84 Fluorinert liquid mixture [9] (the maximum hydrostatic limit is 23 kbar). The cell of the nonmagnetic NiCrAl alloy had an inner and outer diameters of 4.7 and 12.7 mm.

![Figure 5](image.png)

**Figure 5.** Sketch and photo of the clamp pressure cell (HPCC20-NiCrAl) for neutron diffraction studies in magnetic fields and under pressure up to 20 kbar. 1- cell body (nonmagnetic alloy 40HNU (NiCrAl)), 2- nut (nonmagnetic Ti alloy), 3- extrusion ring (CuBe), 4- sample can (Pb), 5- can caps (Pb), 6- pistons (nonmagnetic alloy 40HNU), 7- pistons.

Pistons of non-magnetic NiCrAl were used excepting for the highest pressure for which weakly magnetic pistons of tungsten carbide were used. A magnetic field in the horizontal scattering plane...
was used to induce a controlled magnetic region in the sample, which is especially important at high pressures. The crystallographic axis $[11\bar{1}]$ was parallel to the magnetic field with precision better than 0.2°. The small size of the pressure cell allowed us to thermalize the sample for 15 min in the actual temperature range 1.5-35 K. All $M(T)$ measurements were carried out with increasing temperature and thermal hysteresis was not controlled.

4. High-pressure clamp cell for inelastic scattering on the TOF spectrometer FOCUS

A clamping cell made of a strong Al alloy was developed and tested to perform measurements by inelastic scattering (INS) of neutrons under pressure. The sketch of the cell is shown in Figure 6. The cell is a clamp-type pressure cell, however some features differ from elements with zero matrix pressure. The INS signal is significantly smaller in comparison with an elastic signal, for example, in neutron diffraction measurements. The absorption associated with the cell must be minimized for successful INS experiments under pressure while a sample volumes should be as maximum as possible. The geometry of the TOF spectrometer FOCUS (PSI, SINQ) requires also optimization of the cell geometry in order to minimize data corrections due to anisotropic cell absorption and the presence of a parasitic inelastic intensity arising from elastic neutrons multiply scattered by the sample and the cell. The developed cell meets these requirements. The axial symmetric cell with an internal diameter of 70 mm and an inner diameter of 7 mm was made of a reinforced aluminum type. It provides a sample volume of 2500 mm$^3$ at the maximum pressure of 10 kbar. Aluminum absorbs some few neutrons, but it is not so much strong material. To strengthen the cell an inner tube with a core of 1.5 mm thick, made of strong hardened steel (or an alloy with a zero matrix) was used, and the nut was screwed into a hardened threaded steel insert allowing to support the steel insert (tube) and inner Al-cylinder. Losses due to absorption and elastic scattering on the cell were estimated as about 6 times for the incoming neutron wavelength $\lambda = 2.09$ Å. No inelastic features originating from the cell were observed in the spectra measured for $\lambda = 2.09, 4.3$ and 5.0 Å above and below the Bragg limit. The cell was filled in the same way as for cells with zero matrix. Measuring the INS shift of the pressure calibrator lattice parameter were supplemented by measurements on a powder diffractometer before or after the INS experiment. Experience with clamping cells has shown that the pressure in the cell is maintained for several days and does not depend on repeated thermal cycling.

![Figure 6. Sketch and photo of the clamp pressure cell (HPCC10-Al) for inelastic neutron scattering studies on the TOF spectrometer FOCUS(SINQ) up to 10 kbar ($T = 2$-300K, $V = 1.6$ cm$^3$).](attachment:image.png) 1 - Al alloy B95T, 2 - insert part Al alloy B96T, 3 - inner part steel 45XМНФА, 4 - nuts from steel 45XМНФА.
References

[1] Sadykov R A, Gruzin P L, Suhoparov V L 1995 High Pressure Research 14 199-202
[2] Podlesnyak A, Strassle Th, Mirmelstein A, Pirogov A and Sadykov R 2002 Eur. Phys. J. B 29 547-52
[3] Strassle T, Divis M, Rusz J, Janssen S, Juranyi F, Sadykov R and Furrer A 2003 J. Phys.: Condens. Matter 15 3257-66
[4] Fak B, Sadykov R, Flouquet J, Lapertot G 2005 J. Phys.: Condens.Matter 17 1635-44
[5] Plumier R J 1966 J. Appl. Phys. 37(3) 964
[6] Jun Akimitsu at all. 1978 J. Phys. Soc. Japan 44(1) 172
[7] Kleinberger R and R. de Kouchkovsky 1966 CR Acad. Sc.Paris 262(Ser.B) 628-30
[8] Decker D L 1971 J.Appl.Phys. 42 3239
[9] Sidorov V A and Sadykov R A 2005 J. Phys.: Condens. Matter 17 S3005–8