# 1.1. CONDENSED MATTER PHYSICS

In 1998, at the IBR-2 reactor neutron scattering investigations in condensed matter were conducted using four basic methods: diffraction, small angle scattering, inelastic scattering and polarized neutron optics. During the reported year eight reactor sessions were held. The spectrometer beam time was distributed in accordance with recommendations of Experts Committees on submitted experimental proposals taking into account the existing long-term agreements for cooperation.

In the list of spectrometers operating in the user mode in 1998 there were included 10 instruments: HRFD, DN-2, DN-12, SKAT, YuMO, SPN, REFLEX-P, KDSOG, NERA, and DIN. The new spectrometer in the list is the texture diffractometer SKAT commissioned in spring 1998. It has replaced the spectrometer NSVR. The vertical geometry of the detector system in SKAT makes it possible to improve considerably the conditions of diffraction data registration from coarse-grained geologic samples by increasing, in the first place, the intensity and the effect-to-background ratio. At the diffractometer DN-2 there started experiments with a toroidal high pressure cell for a pressure up to 70 kbar (developed in Institute of High Pressure Research in Troitsk). At the SPN spectrometer possibilities of registration of polarized neutron scattering essentially increased due to new improvements, namely, an adiabatic spin-flipper, a supermirror-based analyzer and PSD were put into operation. In autumn 1998, work started to build a neutron guide for the new Fourier diffractometer FSD. It is planned to put into operation the first stage of FSD next year.

In the 1998 scientific program of the Condensed Matter Division (CMD) traditional research directions prevailed.

In the last three years at IBR-2 the program for investigations of a mercury based superconductor $HgBa_2CuO_{4+\delta}$ (Hg-1201) with different oxygen contents in the basal plane has been conducted. The work consists of precision investigations of the structure of compounds and studies of changes in the structure under the influence of high pressure as $\delta$ changes from 0.06 to 0.19.

HRFD measurements yielded precision structural data, including at low temperatures, and allowed establishing several principle facts at a time. It is shown that the distribution of cations in the lattice corresponds to a stoichiometric distribution without a notable number of vacancies and mutual substitutions and the fact that extra oxygen is only present in the center of the basal plane. The dependence of the superconducting transition temperature on the oxygen content appears to be parabolic with a maximum at $\delta \approx 0.13$ which corresponds to transition in the $CuO_2$ plane of about 60% of carriers formed by oxygen atoms implanted in the mercury plane.

As a continuation of above experiments the atomic structure of a compound $HgBa_2CuO_4F\delta$ where extra oxygen is replaced by fluorine was investigated. The idea of the experiment is to replace a divalent doping element by a univalent one. In this case, maintaining of the charge balance and the ionic nature of carriers formation in the superconducting plane would require a double number of doping atoms to reach an equivalent superconducting transition temperature. The initial phase Hg-1201 with $T_c$=61 K was successfully fluorinated, which first resulted in an increase in $T_c$ to 97 K and then, in its decrease and subsequent suppression of superconducting properties as the fluorine content in the sample increased. Neutron diffraction analysis of two compositions of $HgBa_2CuO_4F\delta$ confirms the insertion of fluorine into the charge reservoir and really demonstrates the doubling of the fluorine content in the structure in comparison with the oxygen phases Hg-1201 with close $T_c$. This fact is a strong argument supporting the ionic model of the formation of electric charge carriers (holes) in Hg-1201 under doping. Also, the fact of the doubling of the fluorine content shows that the apical distance Cu-O depends on the number of anions in the basic plane of the structure and not on their charge. On the contrary, similarly to $T_c$ the distances Cu-O in the plane $CuO_2$ depend on the charge state of the system.

A compound $HgBa_2CuO_{4+\delta}$ has the simplest structure in the family of mercury containing compounds, which makes it possible to investigate small structural changes as a function of applied external pressure. Neutron diffraction investigations of structural changes in a superconducting $HgBa_2CuO_{4+\delta}$ for an optimal doping composition were carried out on the DN-12 diffractometer over the pressure range from 0 to 5 GPa and for compositions with $\delta$=0.06 and 0.19, they were conducted over the range from 0 to 0.8 GPa on the diffractometer D2B (ILL). Compressibility values of elementary cell parameters and certain interatomic bond lengths in the structure were determined. It is shown that the influence of pressure on the interatomic distances strongly depends on the doping level: for $\delta$=0.06, the structure compresses uniformly, in the overdoped state the compressibility of some distances (Hg-O2) decreases to zero and of others (Ba-O3) it increases 2.5 times (Fig. 1). The latter fact points to a considerable change in the concentration of free charge carriers in the conducting planes of $CuO_2$ under the influence of pressure.

In the last years, investigations have shown that close connection exists between

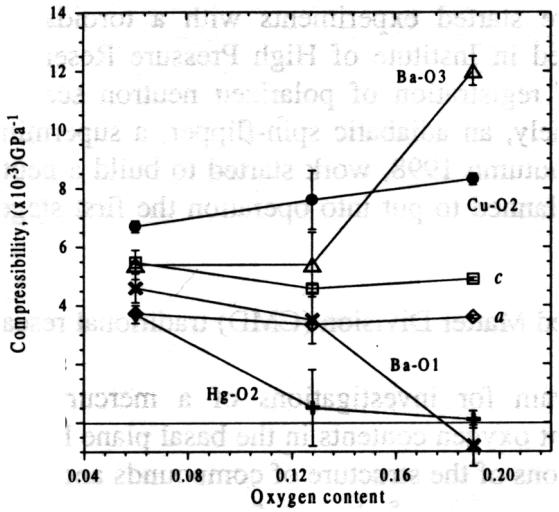


*Fig.1. Compressibilities of the main parameters of the structure Hg-1201 for three doping levels. At a low oxygen content ($\delta$=0.06) the structure compresses almost uniformly. In an overdoped state ($\delta$=0.19) the compression of different elements of the structure is essentially different, which points to the fact that the process of charge transition from the reservoir to the superconducting layer takes place.*

the magnetic and transport (electric) properties of manganites with a perovskite structure, $A_{1-x}A'_xMn^{3+}{}_{1-x}Mn^{4+}{}_xO_3$, where A is La or a rare earth ion, A′ is an alkaline earth divalent cation, Ca, Sr, etc., widely known nowadays as CMR-compounds (CMR means Colossal MagnetoResistance). As a rule, at room temperature such compounds are paramagnetic insulators (PI). At the same time, their low-temperature states are ferromagnetic metals (FMM) or antiferromagnetic insulators (AFI) with a tendency to the ordering of manganese charges (CO). The low-temperature state is determined by the balance of several types of interaction and it can be easily modified by changing the composition of the compound or applying an external action. The realization of any of the states depends on the doping level, i.e., on the ratio $Mn^{3+}/Mn^{4+}$, as well as on the ratio of the bond lengths Mn-O to A-O determined, in the main, by the mean ionic radius of cations $\langle r_A \rangle$ in the A-site of the perovskite structure.

The HRFD structural experiment shows that changes in the transport and magnetic properties of $La_{0.35}Pr_{0.35}Ca_{0.30}MnO_3$ at phase transition are really connected with changes in the atomic structure. Namely, at temperatures below the insulator-to-metal transition temperature taking place in the investigated compound at T≈150 K, there arises the ferromagnetic state and simultaneously, there takes place the "melting" of the orbital ordering of oxygen atoms on bonds (Fig.2), i.e., the equilization of the Mn-O bond distances in the oxygen octahedra.

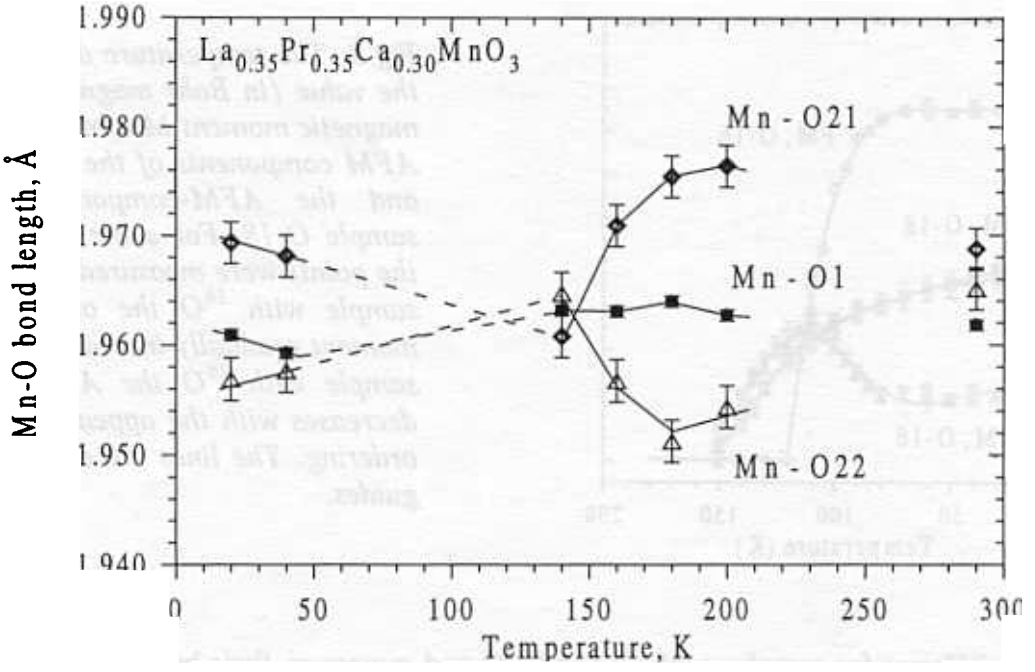


*Fig.2. The dependence of the interatomic distances Mn–O on the temperature. The bond Mn-O1 is directed along the axis b, and Mn–O21 and Mn–O22 lie in the plane (a,c). At T=150 K there arises a saturated ferromagnetic state accompanied with the "melting" of Jan-Teller static distortions of oxygen octahedrons.*

Investigations of the interplay between the electron and phonon subsystems in manganites deserve special attention. In the article by Babushkina et al. (Nature, 391 (1998) 159), the influence of the isotope replacement of oxygen on the transport properties of the compound $(La_{0.25}Pr_{0.75})_{0.7}Ca_{0.3}MnO_3$ are investigated. It appears that because of the critical value of the tolerance factor of the perovskite structure (~0.91) such composition is close to the boundary between the metallic and insulating states , i.e., there develops special sensitivity to different physical effects . As a result, even a relatively small effect, including the isotopic replacement of oxygen, may cause a transition of one state into the other. Namely, it is shown that a compound with a natural oxygen isotope mixture (99.7% $^{16}O$) turns into a metal at T≈110 K while a compound enriched with the isotope $^{18}O$ remains an insulator down to helium temperature.

Later neutron diffraction investigations were conducted on DMC (the neutron source SINQ in P.Scherrer Institute in Switzerland) and HRFD and studied the influence of the isotopic replacement of oxygen $^{16}O$ by $^{18}O$ on the magnetic structure and the charge ordering process in the compound. The evolution of the structure of two samples, one with a natural oxygen isotope mixture and the other enriched with $^{18}O$ to 75% was studied. In the first place, it is established that at room temperature the samples are identical from structural point of view. For example, the coincidence of their elementary cell parameters is within 0.004% from where it follows that the oxygen stoichiometry is not worse than ±0.002. As the temperature decreases, the sample with $^{16}O$ subsequently experiences antiferromagnetic ($T_{AFM}$=150 K) and ferromagnetic ($T_{FM}$=110 K) transitions leading to the establishment of a noncollinear ferromagnetic structure while in the sample with $^{18}O$ there arises a pure antiferromagnetic ($T_{AFM}$=150 K) order (Fig.3). The low-temperature dependence of the diffraction peak intensities connected with charge ordering appear to

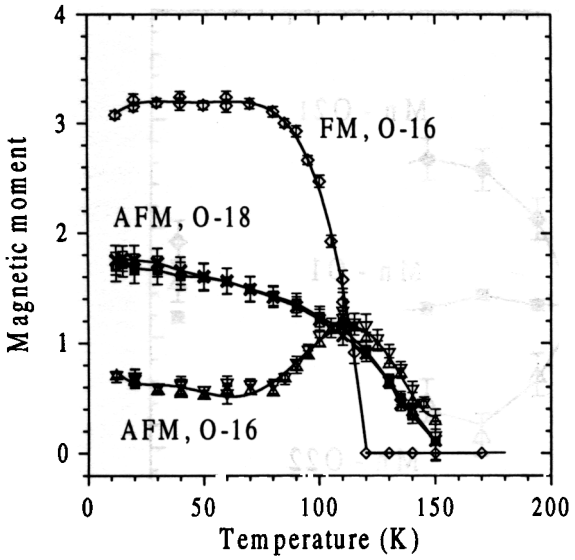


*Fig.3. The temperature dependence of the value (in Bohr magnetons) of the magnetic moment Mn for the FM- and AFM components of the sample O-16 and the AFM-component of the sample O-18. For some temperatures the points were measured twice. In the sample with $^{18}O$ the ordered AFM moment gradually increases and in the sample with $^{16}O$ the AFM moment decreases with the appearance of FM ordering. The lines are drawn as eye-guides.*

be essentially different for samples with $^{16}O$ and $^{18}O$ and moreover, their behavior correlates with the behavior of their electric resistance. The reported experimental results are evidence of the fact that the low-temperature state of $(La_{0.25}Pr_{0.75})_{0.7}Ca_{0.3}MnO_3$ is controlled by strong dynamic electron-phonon interaction due to which the isotopic replacement of $^{16}O$ by $^{18}O$ results in a change of the electron state (metal – insulator) and a totally correlating change in the magnetic structure (noncollinear ferromagnetic – pure antiferromagnetic) (see also Experimental Reports).

An interesting program of investigations in the structure and dynamics of ammonium halides is carried out with the DN-12 diffractometer. For the compounds $NH_4Br$, $NH_4I$, $NH_4F$, $NH_4Cl$ or their deuterated analogous there are obtained data on the equation of state and changes in the structure with regard to pressure as well as the dependence of librational and transverse optical modes on the pressure over the range from 0 to 40 - 50 kbar. The expected increase in the frequency of the two modes as the pressure increased was actually observed but it appeared to be essentially different for different compounds and depended on the type of the mode. Structural data analysis makes it possible to suggest a hypothesis about a single critical value of the position parameter of the hydrogen atom for which there occurs an orientational phase transition of the order-disorder type in all investigated halides. For a detailed report of investigations of inelastic neutron scattering in $NH_4I$ and $NH_4F$ together with details of the application of the Reverse Monte Carlo Method for the analysis of orientational disorder in these compounds see the Experimental Reports Section of this Annual Report.

The small-angle scattering method was used to study an important for applied purposes process of ethanol penetration through a model lipid membrane. The influence of ethanol on the thickness of the lipid bilayer and the intermembrane interaction were studied. Figure 4 illustrates changes in the thickness of a bilayer from DPPC molecules as a function of the ethanol concentration at T=25°C. A decrease in the membrane thickness corresponds to the formation of a phase with interpenetrating hydrocarbon chains. In Experimental Reports detail accounts of the use of the small-angle neutron scattering method in investigation of polymer and lipid systems carried

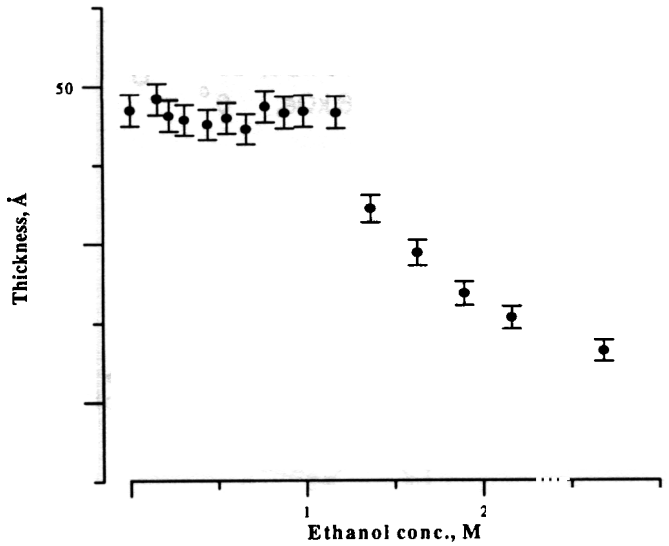


*Fig.4. The dependence of the thickness of the DPPC lipid bilayer on the ethanol concentration at room temperature. A sharp decrease in the thickness at 1.2 M corresponds to the formation of the phase $P\beta_I$ and the beginning of the linear growth of the penetrability of the membrane.*

out in collaboration with specialists from Germany, Slovakia and Czech are presented.

In 1998, the program for the investigation of the ribosome structure continued to proceed in several directions: the structural data on different types of 70S subparticles of the ribosome *T.thermophilus* were obtained by the contrast variation method and the neutron scattering curves on 16S and 21S particles of a small subparticle 30S of the ribosome *T.thermophilus* were measured. Also, above particles were investigated using the method of electron microscopy (see Experimental Reports).

The new methodical possibilities realized on the spectrometer SPN made it possible to start experiments of the observation of an interesting and complicated phenomenon of formation of neutron standing waves on neutron transmission through thin layers. In the first experiments, the total reflection regime and several registration channels of the effect, such as capture γ-quanta or α-particles, spin-flip neutrons, were used. Neutron standing waves with a period from 250 Å to 500 Å were reliably detected (Fig.5) and it appeared possible to demonstrate that their registration allowed the determination of a magnetic noncollinear layer shift inside the medium with an accuracy on the level of 0.1 Å (see also Experimental Reports).

The fundamental issue of the neutron coherence length, i.e. the characteristic dimension within the limits of which the neutron interferes with itself was investigated in experiments carried out on the neutron reflectometers REFLEX (Dubna) and EROS (Saclay). Analysis shows that the experiment of thermal neutron reflection from a homogeneous film on a substrate makes it possible to separate considerably in space two neutron beams and correspondingly, improve the existing estimates of the coherence length. In the time-of-flight experiments the coefficient of neutron reflection from a copper film sputtered on a glass substrate was determined over a wide range of neutron wave vectors. The followed data processing using a function with the neutron coherence length as a parameter allowed obtaining of the lower estimate, namely, $l_{coh} > 5$ mm. This is about 20 times higher than the earlier coherence length estimate and is very important from the viewpoint of correct interpretation of the reflectometry data.

In the recent time, a large series of experiments to measure the vibrational density of states in ammonium and methyl groups of compounds of the type $(CH_3NH_3)_5Bi_2Cl_{11}$ were conducted on the inverted geometry spectrometer NERA. Structural phase transitions in such compounds and in $C_7H_{12}$ crystals and their derivatives were studied by the method of simultaneous registration of the diffraction pattern and inelastic scattering spectra. For polymethylsiloxane oligomers adsorbed on hydrogen-charged or methylated surfaces of amorphous silica, a large volume of factual information

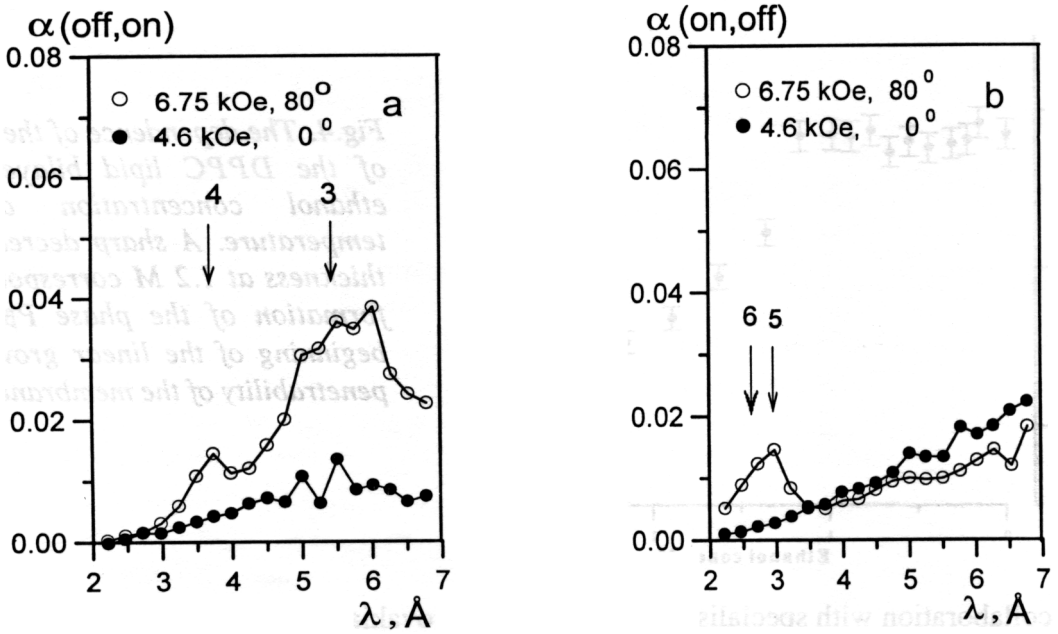


*Fig.5. The curves of the quantities proportional to neutron spin-flip reflection coefficients from the structure Cu(1000 Å) / Ti(2000 Å) / Co(60 Å) / Ti(300 Å) sputtered on glass. The peaks appearing at the wavelengths 3.7 Å and 5.6 Å (lef-side figure) and 2.8. Å (right-side figure) correspond to standing wave antinodes.*

on vibrational spectra of interest for modern nanotechnologies was obtained. In addition, the results of these experiments are used to verify calculations of such systems by quantum chemical methods (see also Experimental Reports).

In 1998, as before, applied research was mainly connected with investigations of internal stresses in volume industrial products on the spectrometers HRFD and EPSILON and studies of rock textures. The spectrometer EPSILON operating in combination with the instrument SKAT

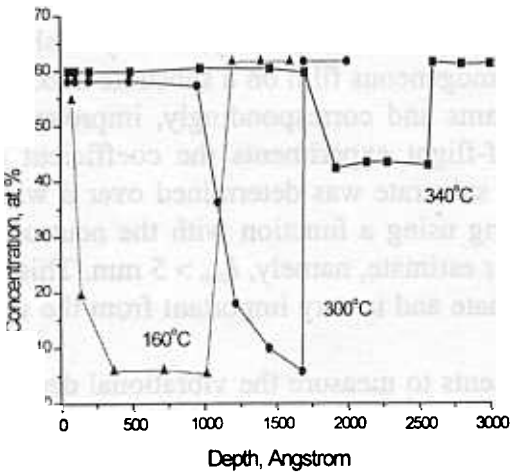


*Fig.6. Oxygen depth profiles for annealed Co films on the glass substrate.*

was created in 1998 by physicists from GeoForschungsZentrum (Potsdam) to study stresses in rocks. The conducted tests show that the resolution of the spectrometer makes it possible to detect internal stresses on the order of 10 – 15 MPa in quartz samples, which is sufficient for the solution of many interesting problems in geology.

On HRFD, investigations of internal stresses in gradient materials continued. In particular, the system CuW was studied in detail and it is discovered that plastic deformation of the sample strongly changes the stresses induced by thermal expansion.

On the spectrometer SKAT, experiments to determine the structure of model calcite samples and measure the velocity of ultrasound waves in them were conducted. The followed calculation based on models taking into account the dependence of the wave velocity on the texturization of the sample yielded values considerably different from the experiment. From here an urgent necessity follows to continue the development of methods accounting for the texture as a characteristic of construction materials of great practical value.

At the EG-5 accelerator, investigations of thin structures used to manufacture neutron polarization mirrors continued. In particular, in 1998 a detail study of the oxidation process of Co and Fe layers on annealing in air in different temperature modes was carried out. The RBS and NRA methods helped to measure depth profiles of the oxygen concentration in films and substrates (Fig. 6). The achieved accuracy of oxygen concentration measurements is 2%. The obtained information will be used in the optimization of the manufacturing process of neutron mirrors.

**Scientific program of the Condensed Matter Physics Division in 1998 was executed in cooperation with the following institutes and organizations**

| | |
|---|---|
| ***Bulgaria*** | University; Institute for Nuclear Research and Nuclear Energy (Sofia) |
| ***Czech Republic*** | Polytechnical Institute (Prague) |
| ***Egypt*** | Atomic Energy Authority of Egypt (Cairo) |
| ***Finland*** | Technical Center (Espoo) |
| ***France*** | Laboratoire Leon Brillouin (Saclay); Institut Laue-Langevin (Grenoble) |
| ***Georgia*** | University (Tbilisi) |
| ***Germany*** | Hahn-Meitner Institute (Berlin); Research Center (Rossendorf); University (Bayreuth); Technical University (Kemnitz); Research Center (Darmstadt); GKSS (Geesthacht); Fraunhofer Institute for Nondestructive Testing (Dresden-Saarbruecken) |
| ***Hungary*** | Research Institute for Solid State Physics (Budapest) |
| ***D.P. Republic of Korea*** | University (Pyongyang) |
| ***Poland*** | Institute of Nuclear Physics (Cracow); University (Poznan) |
| ***Romania*** | Atomic Physics Institute (Bucharest) |
| ***Russia*** | Kurchatov Institute; Institute of Solid State Physics; Institute of Theoretical and Experimental Physics; Petersburg Nuclear Physics Institute; Institute of Physics of Metals; Moscow State University; Institute of Crystallography; Physical Energetical Institute (Obninsk); Institute of Nuclear Physics RAS |
| ***Slovakia*** | University (Bratislava) |
| ***Sweden*** | University (Goteborg) |
| ***Switzerland*** | Paul Scherrer Institute (Villigen); ETH Zentrum (Zurich) |
| ***U.K.*** | Rutherford Appleton Laboratory (Abingdon) |
| ***Uzbekistan*** | Institute of Nuclear Physics (Tashkent) |
| ***Vietnam*** | Institute of Physics (Hanoi) |