precious structural information on these compounds, as for instance the first observation of the increase of the internal distortion of the SiO_6 octahedra with increasing pressure in a powder sample. Furthermore, the data analysis allowed us to identify a set of thermoelastic parameters to constrain the compositional model of the Earth's lower mantle. Assuming that the thermoelastic parameters obtained from this study are applicable to perovskites with moderate iron content, then the comparison of the density and K_T profiles calculated for a mixture of perovskite and magnesiowüstite with those obtained from the PREM [9] model indicates that a pure perovskite lower mantle is very unlikely. On the other hand, a very good match between the PREM density and K_T profiles is obtained for a mixture of 83 vol% $(Mg_{0.93}, Fe_{0.07})SiO_3$ perovskite and 17 vol% $(Mg_{0.79}Fe_{0.21})O$ magnesiowüstite [8]. ■

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DENSITY MEASUREMENTS OF LIQUID IRON ALLOYS AT HIGH PRESSURES: TOWARDS A BETTER UNDERSTANDING OF THE PLANETS

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Physical properties of iron-based liquids are of much interest to better understand both the current state of planetary cores and their formation during the differentiation of planets. Here we present the first experiments performed on metallic liquids in the Fe-Ni-S system, which might be relevant at least to the terrestrial outer-core and the martian core. Using a large-volume press apparatus (a Paris-Edinburgh press), the P-T range of 0-4 GPa and 20-1250 °C was explored by measuring the absorption profiles, hence density, of samples using high-energy x-rays. Equations of state of liquid iron alloys are therefore on the way to be determined, along with accurate melting-phase diagrams as a function of pressure and temperature relevant to geophysical conditions.

GEOPHYSICAL INTERESTS

Density measurements of Fe-based liquids at pressure and temperature relevant to planetary cores are essential to model accurately the core composition and convection. This should help resolve two important geophysical issues: the generation of the Earth's magnetic field and the thermal history of the planet.

Also relevant to these measurements is the differentiation of planets, i.e. at first order, the individualization of a metallic core towards the center of the planet. All these phenomena refer to the liquid state of core materials, which concerns at least the outer terrestrial core, but also probably Mars, Venus and some Galilean satellites such as Ganymede for example, as a substantial magnetic field (roughly a

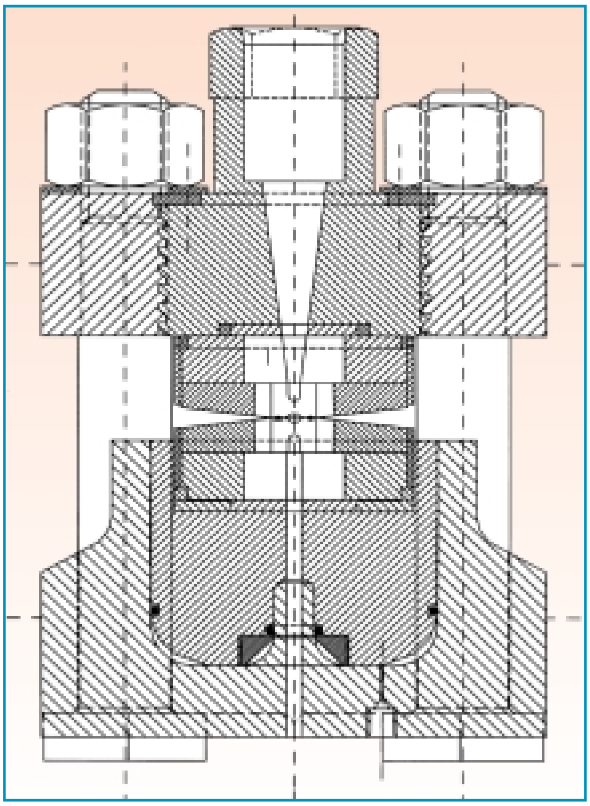

Fig. 1: The Paris-Edinburgh press (model V4). Using a force of 300 tonnes, 300 mm^3 of material (gasket, heater, calibrant, sample...) are compressed so that the maximum pressure of the central 100 mm^3 of sample is 12 GPa. The device weighs approximately 50 kg and measures 20 cm by 20 cm by 25 cm. With high-resistivity graphite heaters, the sample temperature is raised to 1600 °C by passing a current of 100 A through the anvils, giving a maximum power of 250 W.

tenth of the Earth's field in intensity) was recently detected by the NASA's Galileo spacecraft.

Seismic waves, generated by earthquakes, have revealed to be powerful probes of density and elastic properties of the interiors of the Earth, and soon of the Moon and Mars. Indeed, seismic data predict that the terrestrial core is composed of an iron-nickel alloy up to 90 %, plus 10 % of a light element, which is mainly supposed to be either oxygen, silicon, sulfur, or carbon, or a mixture of these elements. The composition of the core can also be assessed using geochemical models of planetary formation, and in fact the consensus which emerges now is that sulfur seems to be quite an ubiquitous component of metallic cores, but in varying proportions (up to 25 wt % for the Martian core for example, and less than 4 wt % in the Earth's core).

The P-T domain of interest differs from one planet to another, ranging from as high as 130-330 GPa and 4500-5500 K for the Earth, to 8-12 GPa and 1500-2000 K for Ganymede. In the experiments discussed below, we have reached a P-T range of 5 GPa and 1200 °C, which is already of great interest for such geophysical and astrophysical problems, even though it needs to be extended to more realistic conditions.

PRELIMINARY RESULTS

Using a large-volume press apparatus (known as the Paris-Edinburgh press - see Figure 1), the P-T range of 0-4 GPa and 20-1250 °C was explored, and high-energy x-ray absorption data (λ = 0.21 Å) were collected *in-situ* using the method developed by Katayama et al. [1] for the study of liquid tellurium. These measurements were carried out on the ESRF High Pressure beamline (ID30) using radiation from the 70 mm period wiggler. The pressure and temperature conditions were determined by computing the intersections of the isochoric lines for hexagonal BN (the sample is contained in a hBN cylinder) and γFe, just before melting. The temperature was also calibrated against the power delivered by the system using these isochoric lines, which allowed us to determine an empirical law for T as a function of the input power, and then to extrapolate it to higher temperatures when the Fe-S alloy was molten.

Disappearance of crystalline diffraction peaks allowed a precise determination of the melting point of samples, while their density was obtained from the x-ray absorption curve (Figure 2) as x-ray absorption obeys the Beer-Lambert law:

$$I/Io = \int_{beam\ size} \exp(-\mu_{liq}\rho_{liq}(X) - \mu_{env}\rho_{env})\, dx$$

where I is the intensity of the x-ray beam, μ the mass-absorption coefficient, ρ the density, and 'env' stands for the sample environment.

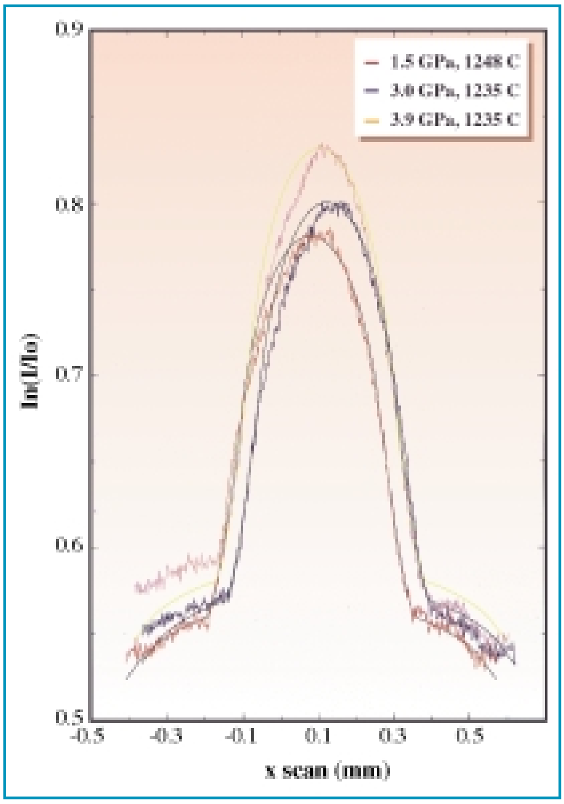

Fig. 2: X-ray absorption scans of 73 % Fe-27 % S liquids. The logarithm of I/Io is fitted according to the Beer-Lambert law (continuous lines, see text) at different pressures (1.5, 3.0 and 3.9 GPa), and around 1250 °C; the background signal corresponds to the sample environment.

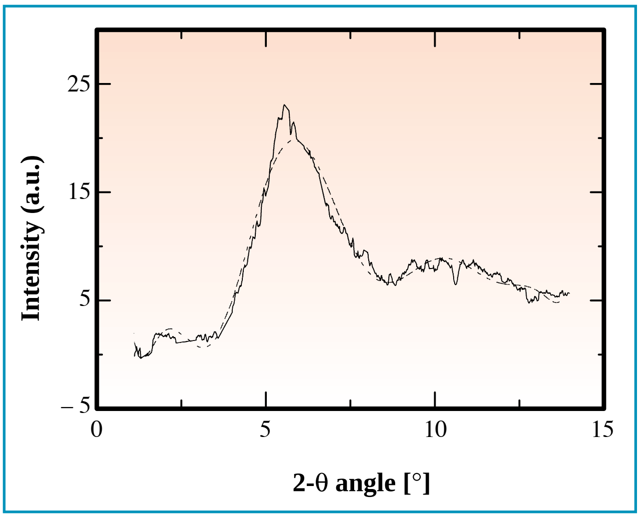

Fig. 3: 1-D integrated diffraction spectra of 73 % Fe-27 % S liquid at 3.9 GPa and 1235 °C. They are fitted with a polynomial function which shows the first and second rings of the radial distribution function.

During our first set of experiments, liquids were obtained in the Fe-S system by mixing Fe and FeS powders to have 25wt % of sulphur. It appears that the presence of sulfur dramatically reduces the bulk modulus, Ko, as Nasch et al. [2] already noticed when carrying out an ultrasonic interferometry investigation of molten Fe-5 %Ni-10 %S at ambient pressure. Indeed, they obtained a value of 63 GPa for Ko, while it is 110 GPa for pure liquid iron, and the value we obtained in these experiments is as low as 27 GPa with 2.5 times their quantity of sulfur. This result could be of considerable geophysical and astrophysical importance, if confirmed in our future studies. In particular, it could be used to determine the maximum sulfur content in the Earth's core, and soon in the Martian core, by direct comparison with core bulk moduli derived from seismological data.

As mentioned earlier, absorption and diffraction data were collected simultaneously, so that in addition to the observation of the disappearance of the crystalline state, the diffraction data gave a first order approximation of the radial distribution function. The integrated diffraction data (the «Fastscan» detector was also used here to collect 2-D images - see article by Fiquet et al. on page 26) of liquid 73%Fe-27%S at 3.9 GPa and 1235 °C (Figure 3) show a broad peak at around 5.5° in 2θ, which corresponds to an interatomic distance of 2.1 Å for the position of the first neighbours, and we can also distinguish a second weaker peak at about 10.5° in 2θ.

Lastly, and of great importance, is the high-precision measurement of the equations of states of the products of metal oxidization. This is carried out using the same high-pressure device, but in a conventional diffraction mode, and also with image plates. Here again the «Fastscan» detector is used, and an example of integrated spectra is shown in Figure 4. Images are collected by first illuminating the sample and its surroundings, then only its surroundings by translation of the whole press, and the difference spectra obtained are of high enough quality to allow analysis by Rietveld refinement. This opens up the possibility of studying the equilibria between liquid metals and solid oxides along planetary P-T profiles. These equilibria are the first order parameters of the establishment and evolution of redox states in the planetary interiors, and these eventually determine the nature of the fluids at the surfaces. Considering the promise of these preliminary results, we are now planning to explore the Fe-Ni-S-O system at higher pressures (up to 8-10GPa) and for various other compositions. ■

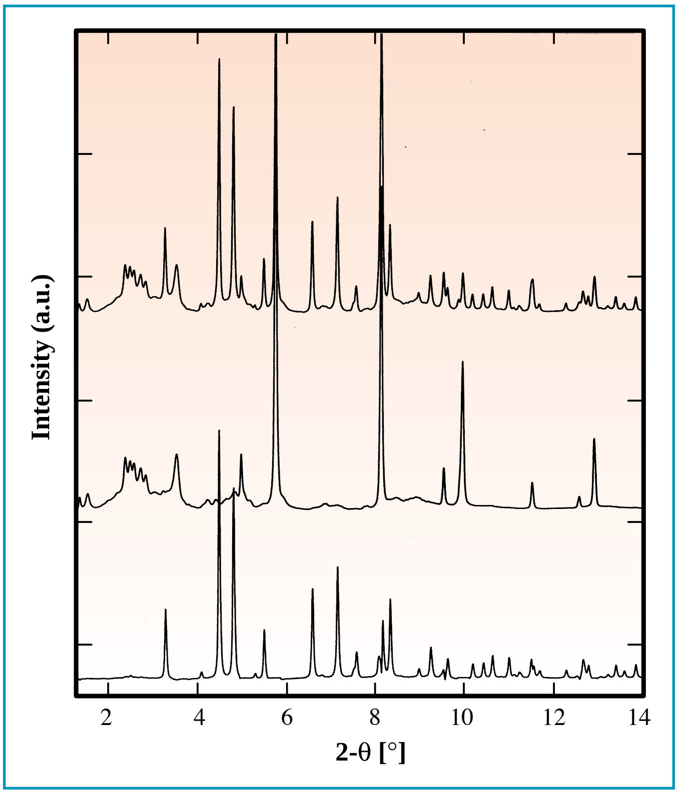

Fig. 4: Diffraction spectra of Fe_2O_3 integrated from 2-D «Fastscan» images obtained in 20 seconds using unfocussed high-energy radiation. Top: sample, calibrant and gasket, middle: calibrant and gasket, bottom: recovered Fe_2O_3 spectrum suitable for Rietveld analysis.

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