New probes for insulators under pressure

J. Mokdad1-2*, C. Marin1-2, D. Braithwaite1-2, G. Knebel1-2 and J.P. Brison1-2

1 Univ. Grenoble Alpes, Grenoble, France
2 CEA-PHELIQS, 38000 Grenoble, France

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*e-mail: Julia.MOKDAD@cea.fr

High pressure has proven to be an extremely efficient tool to tune physical properties in order to reveal and understand the physics of new materials. It is particularly true in strongly correlated systems where the low energy scales involved mean that the ground state of these compounds can be controlled with the application of relatively low pressures. In metallic strongly correlated systems, for example, high pressure can be used to tune a system to a magnetic instability, which may often reveal unconventional superconductivity [1]. For these systems resistivity measurements are ideal as a probe to follow the magnetic ordering temperature. Several years ago we developed resistivity measurements in a diamond anvil cell with an in-situ control of the pressure at low temperature using helium-filled bellows and a mechanical levers [2]. However, there is growing interest in the physics of insulating materials, like Mott insulators or multiferroic systems where pressure is also an ideal tuning parameter. In this case resistivity is not a good physical probe to establish the high pressure phase diagram. We have previously described a system for measuring specific heat with an a.c. calorimetry technique in a diamond anvil cell which can also be applied to insulating materials. However, this technique is not quantitative, and furthermore, the anomaly in the specific heat at some phase transitions may be rather weak. In order to complete this with a more sensitive probe for phase transitions in insulating materials, we describe here the development of a capacitance technique in the diamond anvil cell in order to measure the dielectric constant of insulating materials under high pressure. Furthermore, these systems may be interesting not only at low temperatures but also over a wide pressure range up to room temperature. Maintaining a constant pressure in the bellows when the temperature is swept over a wide range is difficult so we have developed a new in-situ pressure tuning system that relies on a mechanical screw device rather than the helium filled bellows, which may be used over the whole pressure range 2K-300K. Finally, our system also allows to apply a magnetic field up to 9T (see figure 1).

We illustrate the performance of the new device by the determination of the phase diagram of the Mott insulator system GaV₄S₈.

GaV₄S₈ is a Mott insulator of the lacunary spinels family AM₄XS₄ (A=Ga, Ge; M=V, Mo, Nb, Ta; X=S, Se). These systems present very interesting physical properties and very rich phase diagrams at low temperatures and magnetic fields. Many studies exist on GaV₄S₈ at ambient pressure showing a structural transition at 43K and a transition to a ferromagnetic and ferroelectric state at 12.7K, which contains skyrmionic and cycloidal phases at low temperature and low magnetic field. The application of pressure is a very pertinent tool in this family. Applying hydrostatic pressure decreases the interatomic distance and hence increases the transfer interaction while maintaining the same lattice structure [3]. It can be used to control the transfer interaction (t) or the one-electron bandwidth (W) and thus to tune the electron correlation strength. By this procedure, pressure can induce Mott insulator-to-metal transitions. In particular, resistivity measurements under pressure have shown a transition to a metallic phase and even the emergence of superconductivity in some members of the family.

Figure 1: Schematic view of the new in-situ pressure tuning system.

Using our new device and a combination of calorimetry and capacitance measurements, we have determined the evolution under pressure of the temperature of the structural and magnetic transitions and thus established the T-P phase diagram. We find that the structural transition temperature Ts increases with pressure, while that of the magnetic transition stays surprisingly stable. To detect the insulator-to-metal transition, we also follow the evolution of the Mott gap under pressure.
The Mott gap energy shows a linear decrease with pressure which could allow us to predict an insulator-to-metal transition around 14 GPa. (See figure 2)


*Figure 2: The Mott gap energy shows a linear decrease with pressure up to 8 GPa. The pressure of the insulator-to-metal transition could be estimated at around 14 GPa.*