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## **THE AC MULTI-HARMONIC MAGNETIC SUSCEPTIBILITY MEASUREMENT SETUP AT THE LNF-INFN**

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### **Abstract**

AC magnetic susceptibility measurement is a fundamental characterization tool in materials science, to study the dynamic magnetic response of magnetic materials and superconductors. An AC multi-harmonic magnetometer is available at the LAMPS laboratory of the LNF-INFN. The magnetometer allows performing experiments with an AC driving magnetic field with variable amplitude from 0.1 to 20 Gauss and in the frequency range from 17 to 2070 Hz. A DC magnetic field 0-8 T produced by a superconducting magnet can be applied, while data may be collected in the temperature range 4.2-300 K using a liquid He cryostat. The first seven AC magnetic multi-harmonic susceptibility components can be measured with a magnetic sensitivity of  $1 \times 10^{-6}$  emu and a temperature precision of 0.01 K. Here we will describe in detail the magnetometer, the instrumental control and the data acquisition framework. Special attention will be dedicated to the user-friendly LabVIEW-based software platform that allows running the experiments.

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## 1. Introduction

The AC magnetic susceptibility measurement is a suitable method to provide a precise characterization of magnetic and superconducting materials in a non-destructive way, particularly appropriate to study the dynamic magnetic response of the material under investigation [1]. In this experiments the magnetization of the material is periodically changed in response to a time-varying exciting magnetic field  $h(t)$ :

$$h(t) = h_0 \cos(\omega t) \quad (1.1)$$

whose magnetization oscillations can be described as

$$M(t) = \chi' h_0 \cos(\omega t) + \chi'' h_0 \sin(\omega t) \quad (1.2)$$

where  $\chi'$  and  $\chi''$  represent respectively the in-phase and out-of-phase components of the magnetic susceptibility with the applied magnetic field. The in-phase component  $\chi'$  is associated to the dispersive magnetic response of the sample, while the imaginary part  $\chi''$  is proportional to the energy dissipation converted into heat during one cycle of the AC field or the energy absorbed by the material from the applied AC field. Using a complex notation,  $\chi'$  and  $\chi''$  can be combined to form the complex susceptibility. If the system has a magnetic linear response only the fundamental sinusoidal exciting frequency waveform is present. However, when the magnetic response of the sample is nonlinear (e.g., for magnetic or superconductor systems), as a result the pure sinusoidal field induces a distorted waveform characterized by non-sinusoidal oscillations of the magnetization of the material. These oscillations may be described as a sum of sinusoidal components that oscillate at harmonics of the driving frequency:

$$M(t) = h_0 \sum [\chi'_n \cos(n\omega t) + \chi''_n \sin(n\omega t)] \quad (1.3)$$

where  $\chi'_n$  and  $\chi''_n$  ( $n = 1, 2, 3\dots$ ) are the in-phase and out-of-phase components of the harmonic susceptibilities. In the complex notation,  $\chi'_n$  and  $\chi''_n$  can be combined to form the complex harmonic susceptibilities.

$$\chi_n = \chi'_n + i\chi''_n \quad (1.4)$$

As an example, as showed in Fig.1, the anharmonic non-linear magnetization (green line) is the superposition in time of the first and the third components (red and blue lines).

A reliable characterization of materials with a non-linear magnetic response requires the measurement of harmonic susceptibilities beyond the fundamental one. Moreover, the AC magnetic multi-harmonic susceptibility, by probing the real and imaginary parts of the first and the higher harmonic susceptibility, may separate linear and non-linear transport processes occurring in a material, allowing the determination of different magnetic/superconducting phases eventually present in the sample under analysis.

A multi-harmonic AC magnetic susceptibility measurement setup is available at the LAMPS laboratory (*LABoratory for Magnetism High Pressure and Spectroscopy*) of the Laboratori Nazionali di Frascati (LNF) of the Istituto Nazionale di Fisica Nucleare (INFN). This accurate measurement system controls different instruments with different software packages. To measure the susceptibility we need to change the temperature thousands of

times during an acquisition and it is almost impossible to record data and display them using the original instrumental controls and the data acquisition packages. In order to achieve an optimized management of all instruments and a full computer controlled data acquisition, display and recording, a dedicated software platform has been developed.

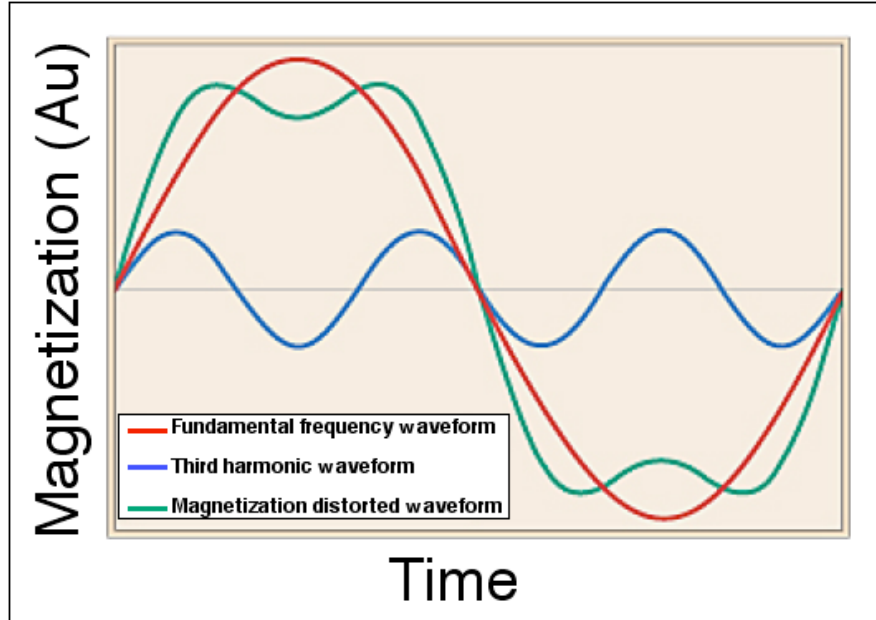


Figure 1. Schematic illustration of the alternating field response of an ideal material with a nonlinear magnetic response.

As a graphical programming language, LabVIEW (National Instrument's Laboratory Virtual Instrumentation Engineering Workbench) has become a very popular development tool widely used in many scientific research and industrial areas. The easy availability of hardware drivers for a very large number of scientific instruments, the easy-to-use multithreaded programming, the convenient graphic user interface (GUI) design, the high-efficiency debugging functions and many other remarkable features make LabVIEW an ideal software development tool particularly optimized for an instrument-oriented programming environment. Actually, it has been also already successfully used for other material magnetic measurement systems [2-4].

In this note, we will mainly describe the schematic diagram and the working principle of the AC magnetic susceptibility measurement setup available at LAMPS. We will also introduce the control of the instruments, the data acquisition framework and the user-friendly LabVIEW-based software platform. Finally, as an example we will present the results of measurements performed on a superconducting sample of the  $N_dFeAsO_{1-0.14}F_{0.14}$  pnictide obtained using this multi-harmonic AC magnetic susceptibility setup vs. temperature.

## 2. The measurement setup and the working principle

In this AC magnetic susceptibility setup the magnetic moment and the susceptibility of the sample are measured by the induction in two counter-wound coils while the AC-field is applied. The voltage induction in the coils is directly related to the magnetic moment in the

sample. Fig.2 illustrates a schematic layout of this AC magnetometer. The heart of the instrument is a gradiometer based on a bridge made by two pick-up coils connected in series and wound in the opposite sense. It is surrounded by a driving excitation coil, which is called the first derivative configuration of the gradiometer coil. The design is used to reduce the magnetic field fluctuations noise in the detection circuit due to the applied magnetic field. The excitation coil receives the AC signal from a function generator, magnified by a custom made amplifier, that yields to an alternating driving magnetic field, whose frequency may range from 17 Hz to 1070 Hz with a variable amplitude from 0 to 20 Gauss.

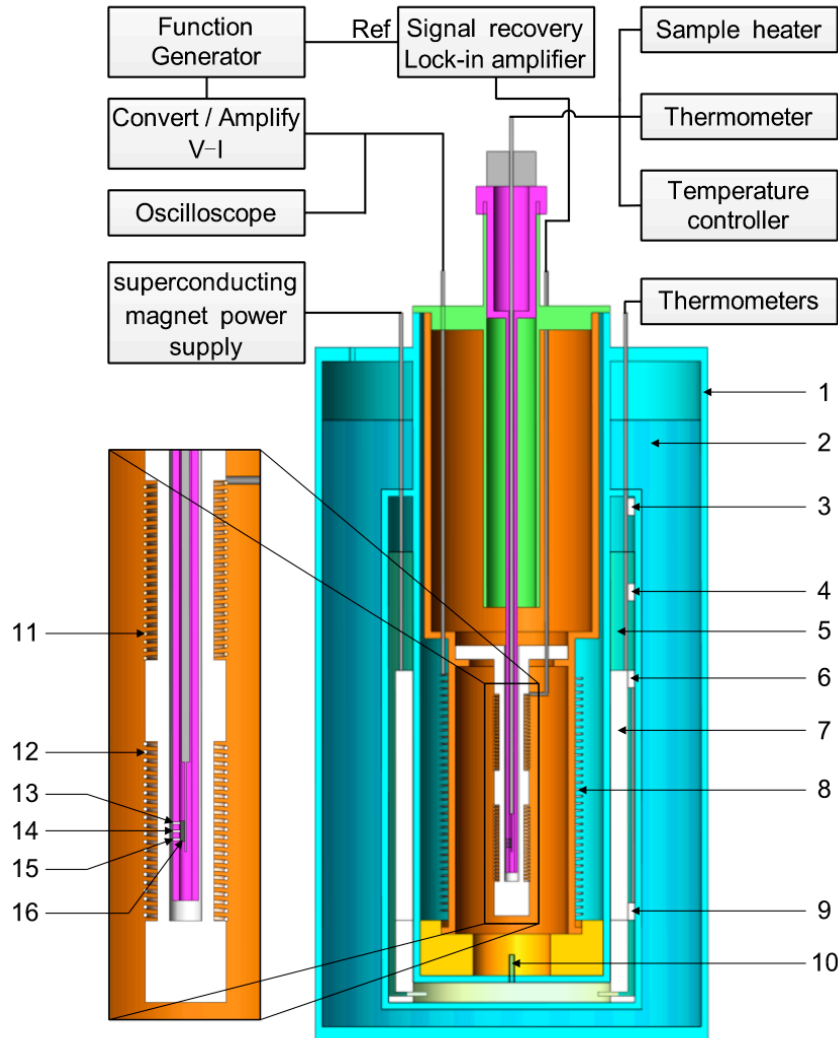


Figure 2. Layout of the AC magnetic susceptibility measurement setup: 1. double vessel cryostat, 2. liquid  $N_2$ , 3. carbon resistor inside the chamber, 4. carbon resistor at the VTI position, 5. liquid He, 6. carbon resistor on top of the superconducting magnet, 7. superconducting magnet, 8. excitation coil, 9. carbon resistor at the bottom of the superconducting magnet, 10. pinhole, 11. pick-up bridge coil (balance coil), 12. pick-up bridge coil (sensing coil), 13. Pt thermometer, 14. carbon resistor thermometer, 15. carbon resistor (sample heater), 16. sample.

The sample is mounted on a sapphire holder slab sets at the center of one of the two pick-up coils (the “sensing” coil) of the bridge while the second one (the “balance” coil) has to remain empty. The sapphire is chosen as a substrate because although being non-magnetic is a good thermal conductor, and it is also characterized by a low electric conductivity that guarantees low current losses. A Pt thermometer and a carbon resistor placed near the sample are in thermal contact with the sapphire holder. The first measures the temperature of the sample while the carbon resistor is used to heat the sample in order to perform experiments vs. temperature. The operational temperature range of this instrument goes from 4.2 to 300 K. The AC magnetic field generated by the driving coil induces a variable magnetic moment on the sample and consequently a flux variation in the pick-up coils, whose voltage signal is measured by a multi-harmonic lock-in amplifier. The lock-in amplifier acts as a discriminating voltmeter. It measures the amplitude and the relative phase of the AC signal, using the same fundamental frequency of the excitation as the reference signal while a fixed phase relationship is provided respect to it. The lock-in amplifier is a band pass filter with a very large Q with its center frequency sets at the selected signal frequency. The output is an amplified DC voltage proportional to the synchronous AC input signal.

The sample holder and the coils assembly can be cooled. Indeed, they are located in a double vessel thermally controlled He gas-flow cryostat. The outer vessel is filled with liquid nitrogen, while the liquid He occupies the inner one, where a superconducting magnet may operate in the range from 0 to 8 T in persistent or non-persistent modes. The sample is mounted at the center of the superconducting magnet and its temperature can be changed via a cold gas flow of He from a pinhole that is manually controlled via a throttle and a needle valve on the top of the cryostat. Before any measurement a purging of the sample compartment with clean He gas has to be done.

The magnetic measurements can be performed both in zero field cooled (ZFC) and in field cooled (FC) modes. In the ZFC mode the sample is slowly cooled below the transition temperature without the DC magnetic field, then the magnetic field is turned on, while in the FC mode the magnetic field is turned on above the critical temperature  $T_c$  of the superconducting or of the magnetic phase. Then the sample is cooled down below the transition temperature and after that, the temperature is let to increase and the measurement starts.

AC magnetic multi-harmonic susceptibility experiments with a magnetic sensitivity down to  $1 \times 10^{-6}$  emu and a temperature precision of 0.01 K can be performed with this setup. Higher components of the magnetic susceptibility can be collected up to the seven harmonic. All these characteristics make the instrument particularly suitable to probe weak magnetic phases present in diluted magnetic structures, as antiferromagnetic, super paramagnetic, spin glass systems and the transport properties of many complex materials and in particular, to investigate the vortex dynamics of superconductor materials.

### **3. Instrumental control and data acquisition structure**

Fig. 3 shows the instrumental control and the data acquisition structure of the AC magnetic susceptibility measurement setup. We utilize the HP 8116A Function generator to generate a sinusoidal signal, which is magnified by a custom made amplifier. This sinusoidal

signal is monitored by the Tektronix TDS 1002 oscilloscope, which also transfers it to the excitation coil.

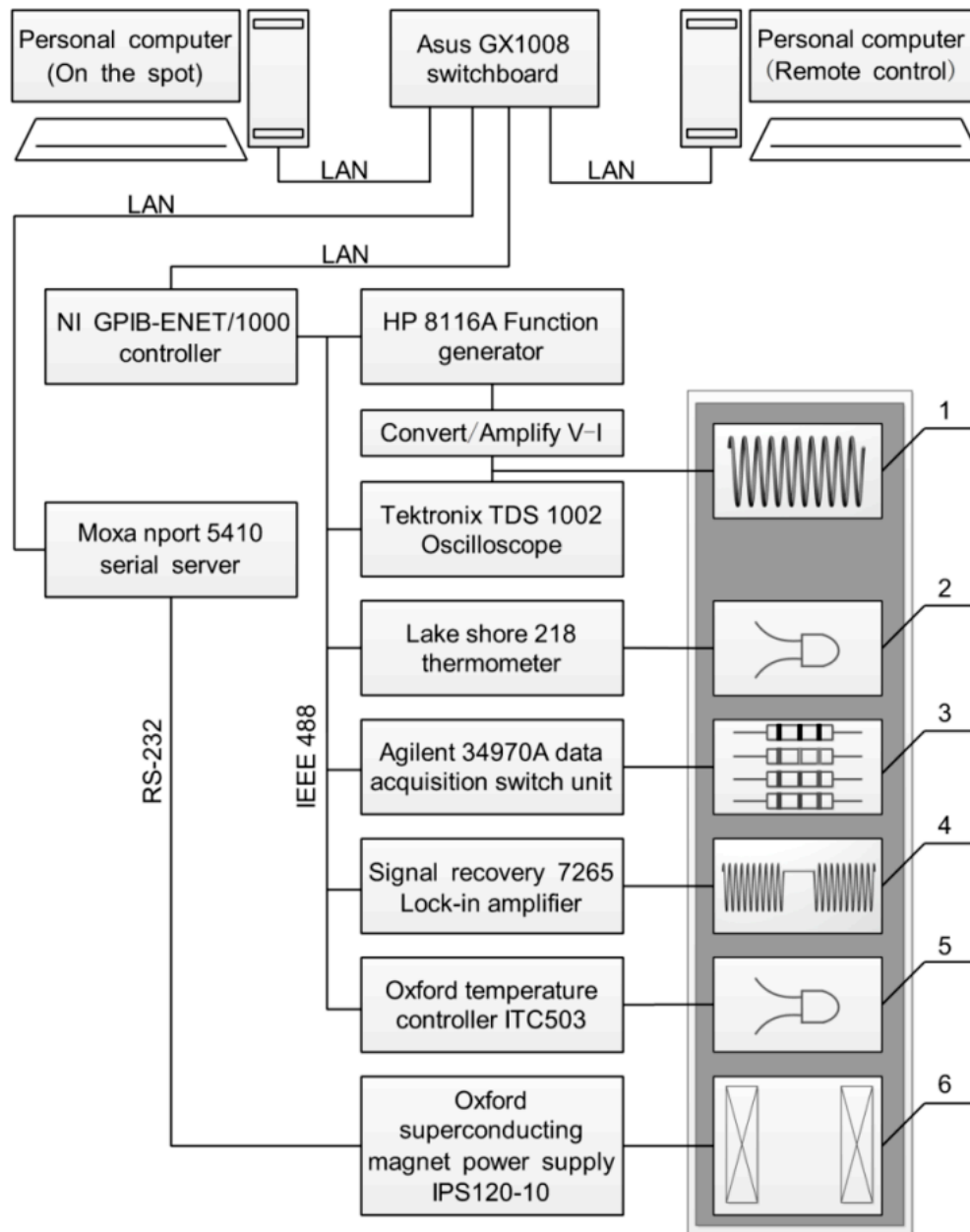


Figure 3. Control and data acquisition logic of the magnetic susceptibility measurement setup: 1. excitation coil, 2. Pt thermometer, 3. carbon resistors, 4. pick-up bridge coil, 5. carbon resistor, 6. superconducting magnet.

The signal recovery 7265 multi-harmonic lock-in amplifier is used to measure the voltage signal generated by the flux variation in the pick-up coils. The Lake Shore 218 thermometer and the Oxford temperature controller ITC503 both collect the temperature of the inspected sample. The DC superconducting magnet is controlled by the Oxford

superconducting magnet power supply IPS120-10. An Agilent 34970A data acquisition switch unit is dedicated to measure the resistance value of the four carbon resistors mounted at four reference positions of the liquid He tank, by which we could evaluate continuously the level of the liquid He inside the reservoir.

Communications of all instruments and their management with a personal computer are realized in different ways following the correlated protocols, as schematically illustrated in Fig.3. The configuration includes two personal computers, one located near the instruments and the second located in the control room. Both should be capable to run data acquisition and to control instruments. The mission is fulfilled by means of special network communication devices and the help of a serial server MoxaNport 5410. Moreover, the original RS-232 serial communication mode of the superconducting magnet power supply was transferred to the Ethernet-based (LAN) TCP/IP protocol making also this instrument available on the network. A high-performance Ethernet-to-GPIB controller (NI GPIB-ENET/1000), with a maximum GPIB transfer rate of 5.6 MB/s supports the access via TCP/IP to all aforementioned instruments. Finally, the Asus GX1008 is the switchboard that allows sharing all instruments among users on the network by hinging together the serial server, the Ethernet-to-GPIB controller and two personal computers.

#### **4. The LabVIEW-based software platform**

When starting to develop a software platform, at first we have to achieve using LabVIEW both instrumental control and data acquisition for each instrument. Only after we started to deal with the software framework based on desired measurement procedures. Typically, every programmable test and measurement instruments have a set of commands that may be controlled by LabVIEW. Actually some commands such as the identify command are the same for most of the instruments. However, for the majority of the instruments, commands and parameters are unique and even change among models of the same manufacturer. Luckily, the LabVIEW drivers greatly simplify the work by abstracting at the lower level the commands of each instrument and providing a familiar API (Application Programming Interface) for all instruments.

Fig. 4 and Fig.5 are the program flow chart of the LabVIEW software that controls instruments and, respectively, the GUI of the AC susceptibility measurement software platform. In the next we will describe how the system is made and how run experiments. After starting the program, users need to log in from the primary menu with a username and a password. Help file and contact information are also accessible from the menu. In the second phase users would be asked by a two buttons dialog to enable the superconducting magnet field. The positive answer starts the superconducting magnet controlling sub-function, while the program jumps to the next procedure if you likely run experiments with the DC magnet field sets to zero. In the following step, users are required to set experimental parameters and instruments: file name of the recorded data, measurement time interval, a valid temperature range for the sample. When the temperature of the sample changes beyond the defined range, the program automatically shut down and a buzzer alarm sounds to warn users of the end of the experimental run. On the right part of the GUI we found how to set each instruments. We may also write notes concerning the experiment in the bottom of the GUI. They will be stored in a text file.

After the setting of the required parameters is completed, the "Start Acquisition" button triggers the initialization procedure and all devices involved in the acquisition can be initialized. After the run started the temperature controller, thermometer and signal recovery acquire data and display them as a graph temperature vs. time, together with the 1<sup>st</sup>harmonic and the other harmonics vs. temperature indifferent pages of the Tab control. Data will be stored with a featured format as a text file that will allow an off-line data processing by users. From the main GUI of the software platform, sub-functions devoted to the measurement of the level of the liquid He and to the excitation signal waveform monitoring are available via the "He detection" and "Oscilloscope" menu, respectively.

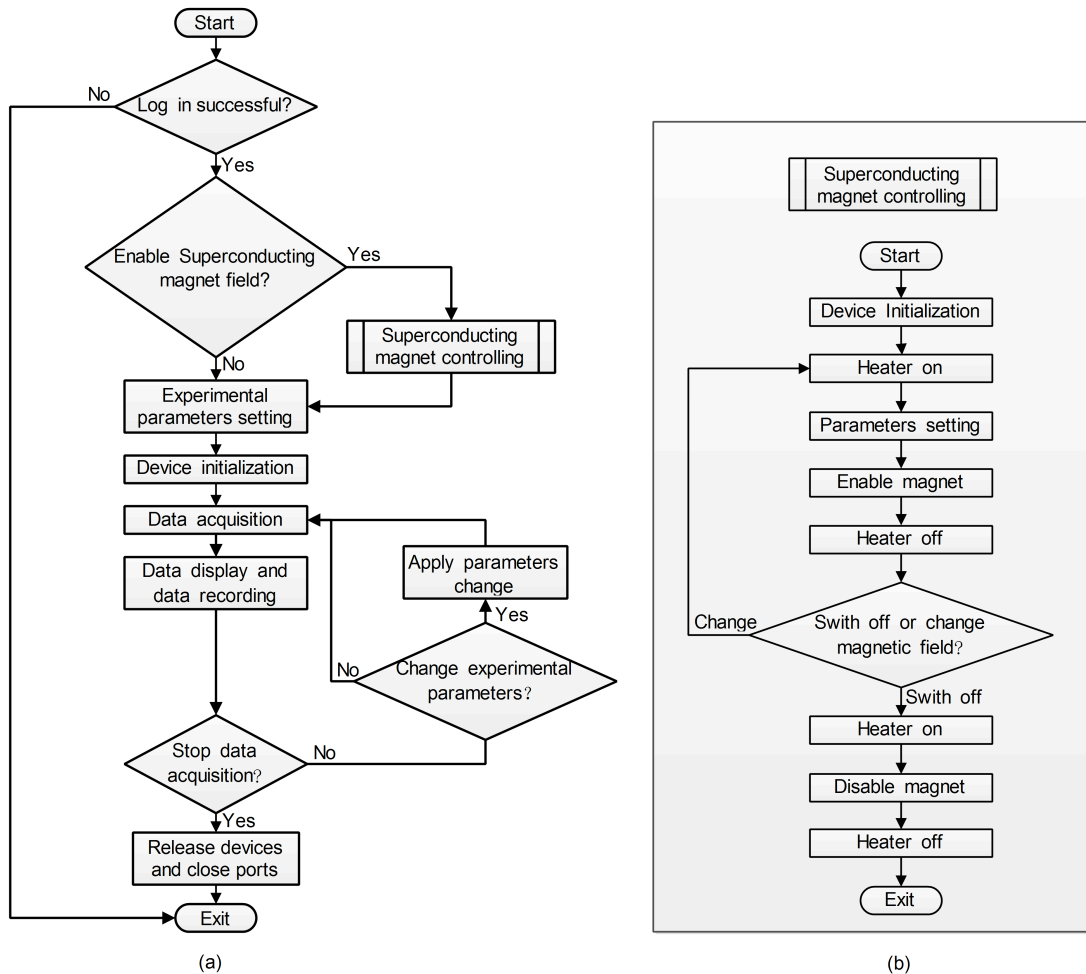


Figure 4. Flow chart of the (a) main function, and (b) superconducting magnet control system.

Fig. 6 is the GUI of (a) the superconducting magnet control, (b) the waveform of the excitation signal monitoring and (c) the liquid He level measurement in the tank. When performing experiment in the DC magnet field environment, the superconducting magnet control function appears and the magnet field would be enabled by specific procedures, whose flow chart is showed in Fig.4(b). To check the sinusoidal signal transmitted to the excitation



coil, waveform monitoring of the excitation signal is usually performed at the beginning of a measurement.

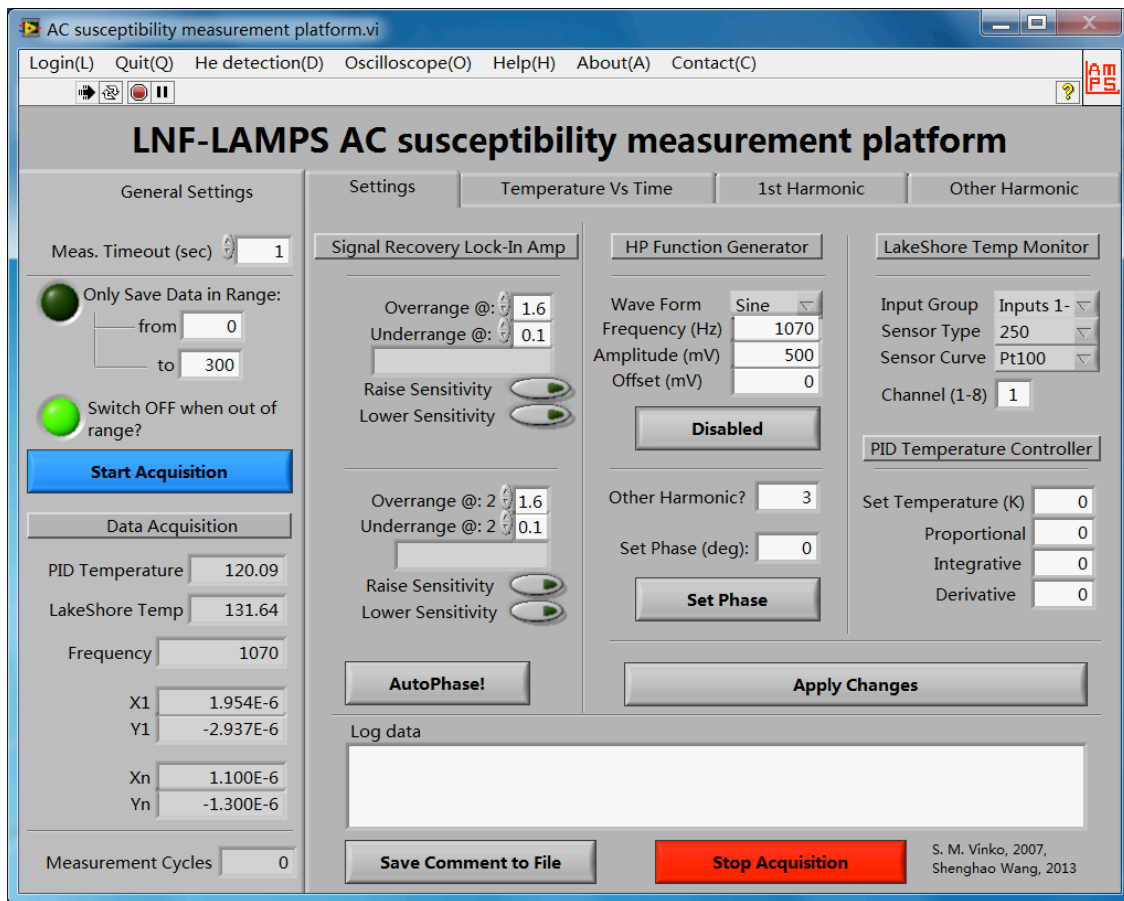


Figure 5. GUI of the AC susceptibility measurement software platform.

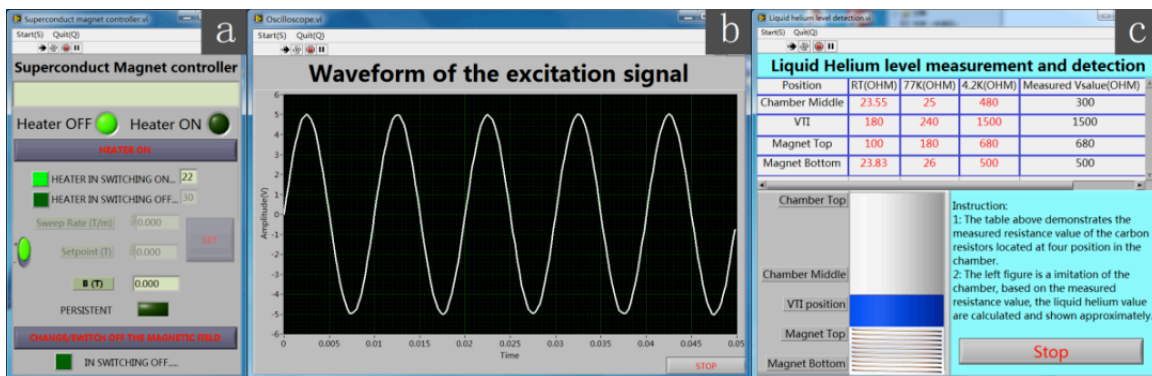


Figure 6. GUI of (a) the superconducting magnet control; (b) waveform of the excitation signal monitoring and (c) measurement of the liquid He.

The measurement of the liquid He level works as follows. The upper portion of the GUI is the chart of the reference resistance values of the four carbon resistors (located at four different vertical positions: middle of the chamber, the VTI position, the magnet top and

bottom) at room temperature, 77 K and 4.2 K, respectively, along with the measured value acquired by the Agilent data acquisition unit. The left below container simulates the liquid He tank, based on measured and reference resistance values of each resistor, so that the level of the liquid He can be evaluated and shown in real time. During experiments with the superconducting magnet field on, the level of the liquid He is gradually reducing and, to avoid the damage of the magnet we need to check that the magnet is fully immersed in the liquid He. This software can be then used to continuously monitor the level of the liquid He. Once the liquid level falls below the top position of the superconducting magnet, the computer will sound a buzzer alarm to warn users to shut down the magnet and abort the experiment. Also this sub-function is used to judge whether the liquid He fills completely the tank during are topping up procedure.

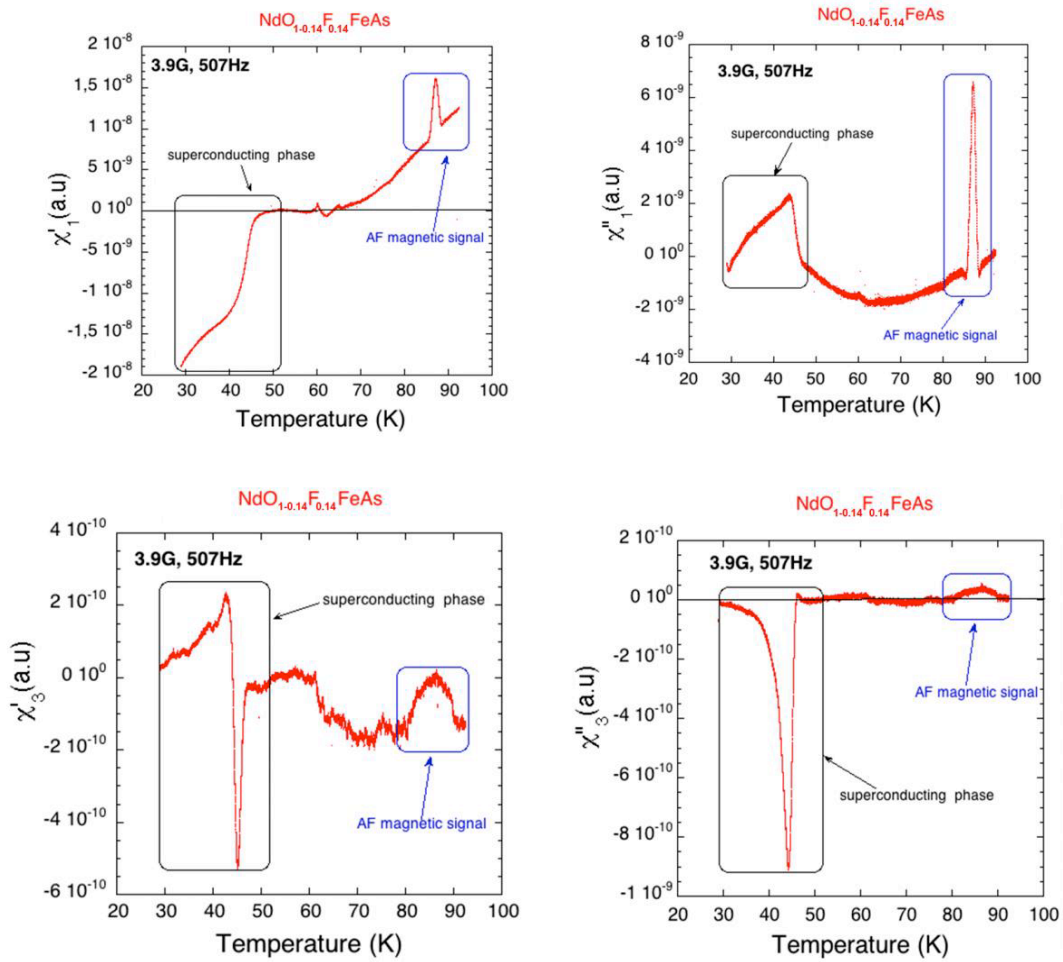


Figure 7. AC multi-harmonic susceptibility measurements of the  $\text{NdFeAsO}_{1-0.14}\text{F}_{0.14}$ : (top): real ( $\chi'_1$ ) and imaginary part ( $\chi''_1$ ) of the first harmonic; (bottom): real ( $\chi'_3$ ) and imaginary part ( $\chi''_3$ ) of the third harmonic.

A software installation package has been generated by LabVIEW with all the aforementioned customized functions and then installed on the two personal computers of the LAMPS laboratory, leaving only shortcuts to users of this susceptibility device. In this way, the source code is protected by accidental changes from users during experimental runs.

## 5. Experimental results and discussion

To give a better feeling of the instrument we show in the last figure data of AC multi-harmonic susceptibility measurements performed on the  $\text{NdFeAsO}_{1-0.14}\text{F}_{0.14}$  compound using the experimental setup of Fig.2. The LabVIEW-based instrumental control and the data acquisition software platform are schematically described in Fig.3 and Fig.4, respectively. The sample characterized was synthesized in Beijing (China) by a high-pressure synthesis method from Nd, As, Fe,  $\text{Fe}_2\text{O}_3$ ,  $\text{FeF}_3$  powders. Additional information of this sample are available in Ref. [5]. For the experiments the sample was cooled with a zero field cooling (ZFC) procedure, the amplitude of the applied AC magnetic field was set to 3.9 G and data were collected at the frequency of 507 Hz. First and third harmonics of the AC magnetic susceptibility were recorded and data are shown in Fig.7. From the behaviour of the first and third harmonics of the AC magnetic susceptibility vs. temperature we can recognize a superconducting diamagnetic phase around  $T_c$  (47 K) and an antiferromagnetic phase around  $T_m$  (90 K). They are clearly separated, in particular in the plot of the real part of the first harmonic  $\chi'_1$ . Moreover, the imaginary part of the first harmonic  $\chi''_1$  shows an increase of the area around the phase transitions. Both the real and the imaginary parts of the third harmonic, named  $\chi'_3$  and  $\chi''_3$ , respectively, confirm that both phase transitions occur. The measurement points out also that in the  $\text{NdFeAsO}_{1-0.14}\text{F}_{0.14}$  compound the anti-ferromagnetic and the superconducting phase coexist, a phenomenon unusual in a superconductor material. This observation could be explained in this sample by the presence of an exchange field of local Nd  $4f$  spins (AF phase) and  $3d$  Fe electron pairs in the FeAs layer (superconducting phase) of this pnictide system [6].

## 6. Conclusion

In this manuscript we describe in detail the AC magnetic susceptibility measurement setup available at the LAMPS laboratory of the INFN-LNF, its instrumental control and the data acquisition framework. This system is running since 1998 and obtained many scientific achievements [7-12]. In this contribution a special attention has been devoted to describe the LabVIEW-based user-friendly software platform used to run the experimental acquisitions with this setup.

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