Magnetic transition of ferromagnetic material at high pressure using a novel system

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A system for the investigation of the magnetic properties of materials under high pressure is fabricated based on diamond anvil cell (DAC) technology. The system is designed with an improved coil arranged around the diamond of a non-magnetic DAC. Using this system, the magnetic transition of ferromagnetic (Fe) sample under increasing pressure can be observed. We successfully obtain the evolution of magnetic properties as a function of applied pressure reaching 26.9 GPa in the Fe sample. A magnetic transition is observed at approximately 13 GPa, which is consistent with the theoretical prediction.

Keywords: diamond anvil cell, magnetic transition, mutual inductance method, transition pressure

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1. Introduction

Diamond anvil cell (DAC) provides an effective and reliable method to obtain ultrahigh static pressure on samples under observation.[1–3] High-pressure research has been advanced by combining DAC with other techniques such as high-energy synchrotron X-ray diffraction,[4,5] Raman scattering,[6–8] and *in situ* resistance measurement.[9,10] Several properties of matter under high pressure can be detected and analyzed using these measurements. Nevertheless, the effect of pressure on magnetic property is rarely mentioned.

Magnetism is not only a macroscopic physical parameter, but also closely related to atomic and crystal structure as well as other microstructural aspects. In addition to high-pressure physics, geology, materials science, and other relevant disciplines have put forward some theoretical predictions on the magnetism rule under pressure.[11–14] Numerous experiments in which the indirect observations, such as X-ray methods[15] and Mössbauer techniques are used,[16–18] have been performed to validate and analyze the results obtained, and the relevant theoretical studies have also been conducted. Since the 1980s, DAC-based magnetic measurement technology has already been explored in some laboratories. One of the most promising methods is the alternating current (AC) susceptibility measurement technology.[19–21]

A similar technology has already been used to observe the effects of pressure on the values of transition temperature $T_c(P)$ of superconductors and ferromagnetic metals, and impressive results have been achieved.[22–26] The reformative systems show that the sensitivity of AC susceptibility can be increased by some approaches, including the geometrical configuration modification of the coils,[27–31] an additional modulating low-frequency magnetic field,[32–34] and improved relevant circuits.[29]

However, *in situ* measurement of magnetic permeability in DAC under extreme compression is still difficult. One of the limitations arises from the extremely small sample volume, which implies a poor filling factor of the induction coil.[30] Furthermore, the magnitude of the background signal, which is influenced by the position modification of the DAC metal parts, changes when a pressure is applied. Nonlinear variation in the background signal prevents the separation of the sample signal from background signal and hinders the achievement of a sufficient sample ratio of signal to background signal.

Although the analysis of the effect of pressure on susceptibility signal transition temperature T_c (P) is important in research, the influence of pressure on magnetic property is also crucial. Investigating the pressure-induced magnetic property transition and measuring the transition pressure at constant temperature P_c are more intuitive and helpful in understanding the effect of pressure on magnetic property of material.

To obtain a direct measurement of magnetic transition pressure P_c, a new system and method are designed. The magnetic permeability variation of a material under pressure is realized. Using this system, the pressure-induced magnetic transition of an Fe sample at room temperature is successfully observed according to the theoretical prediction.

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2. Design of measure system

The basic principle of the developed measurement system of pressure-induced magnetic signal variation is shown in Fig. 1, where number 4 is the measured sample under pressure, and numbers 1–3 represent the detector consisting of three coils inside the DAC, each with the resistive component and inductance component. The excitation signal of the detector is provided by the internal oscillator of the lock-in amplifier.

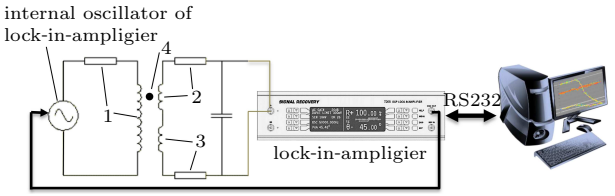


Fig. 1. (color online) Schematic diagram of the recording system for the investigation of magnetic properties under high pressure: 1 exciting coil and its resistive component; 2 detection coil and its resistive component; 3 compensation coil and its resistive component; and 4 sample under high pressure.

Two coils with the same specifications but opposite directions, namely, the induction coil 2 and compensation coil 3, are arranged in the magnetic permeability measurement device. Theoretically, when the parameters of the two coils are the same and the excitation coil is arranged in the desired position, the output signal would approach to zero. However, in practice, many uncontrollable or human-induced factors affect the production and arrangement process of the coils. Thus, the influence of errors must be minimized through structural optimization. Several former arrangements are combined to redesign the structure of the detector (Fig. 2). The new design occupies smaller space and is easier to adjust to the desired position, which reduces the difficulty in detector processing, thereby increasing the production of the finished products.

Figure 2 is the actual layout of the designed detector in the DAC. The volume of the tested sample 6 within DAC is very small. The sample chamber is 180 μm in diameter and 70 μm in thickness. Although the diameter and thickness of the detection coil 2 are reduced as much as possible, the signal produced by the sample of such a small volume in the detector (1–3) remains in the μV magnitude. To identify weak signals of this magnitude, the effects of the inevitable mechanical and temperature changes on the background signal of the detector during the experiment must be excluded. The main factors that affect the background signal are as follows: the stability of the excitation signal, the magnetic signals of other substances near the detector, the eddy current signals inside the sample generated by the excitation signals near the detector, the symmetrical precision of the coils inside the detector, and the connection reliability of the interfaces in the signal circuit. Coils in the detector are fixed using non-magnetic pouring sealant on the support, with each end of the coil connected to the lock-in amplifier through the circuit board 7. The system includes a specific self-written software. The settings of the lock-in amplifier are controlled, and experimental data are collected according to predetermined steps.

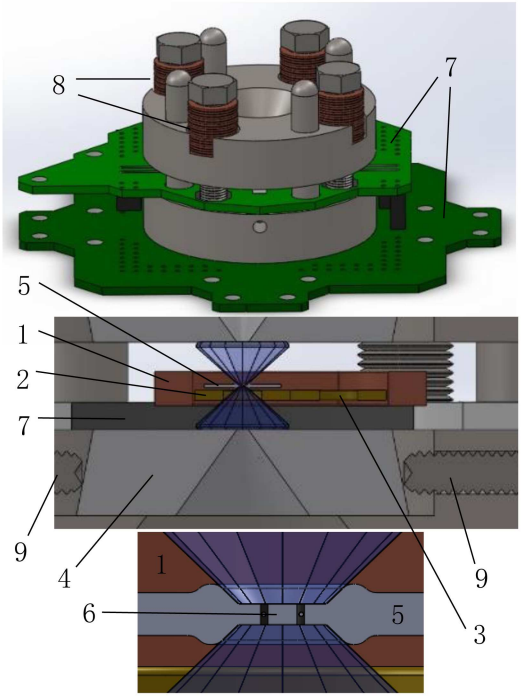


Fig. 2. (color online) Schematic view of the experimental setup. 1 Exciting coil, 2 detection coil, 3 compensation coil, 4 supports, 5 gasket and sample chamber, 6 sample under study and ruby for determining the pressure, 7 circuit boards, 8 belleville springs, 9 set screws, and 10 pressure bolt. All parts are non-magnetic. The structure of the detector has been redesigned. Circuit board can not only work as jig and fixture, but also transmit the signal.

The magnetic permeability of the hard alloy that is widely used in DAC is almost of the same order of magnitude as that of the sample. Given that the volume of the alloy is significantly larger than the sample size, the introduced interference is greater than the sample signal. The common solution to this problem is to use a non-magnetic Be–Cu alloy with good temperature characteristics in place of the traditional ferromagnetic alloy.[29,33,35,36] In the measurement with constant pressure but with varying temperatures, the temperature performance of Be–Cu meets the requirements. Be–Cu has a very good electrical conductivity. The eddy current generated under the influence of the AC excitation signal is stronger than those in other materials and is of the same order of magnitude as the sample signal, thereby causing serious interference to the measurement of sample signal. When measuring the magnetic signal as a function of pressure under isothermal conditions, the metal members arranged in the vicinity of the detector need to meet the following conditions: sufficient hardness and toughness, non-magnetism, low electrical conductivity, and sufficient mechanical machining accuracy. The titanium alloy is selected as the material for the main structure, pressure bolts 10, and other components of the DAC. The quenched and tempered non-magnetic wolfram carbide alloy

is used as the support 4, and tungsten with a purity of 99.9% is used as the gasket 5. Belleville spring 8, set screws 9, and other DAC parts are also non-magnetic. Experiments prove that the conditions are completely satisfied.

3. Experiment

The induction coil 2 and the compensation coil 3 used in the experiment are made of pure Cu-enameled wire with a diameter of 40 μm, an inner diameter of 2 mm, and a thickness of 0.5 mm. Each coil has 180 turns. The excitation coil is made of constant enameled wire with a diameter of 800 μm winding over the outboard with two other coils (2 and 3). The excitation coil is 2 mm thick and has a total of 300 turns.

Fe is selected as the sample to verify the device performance in the experiment. Commercially available 99.9% purity Fe sample was loaded into a gasketed high-pressure DAC (Fig. 2) with a 400-μm diameter culet. The 99.99% purity wolfram gasket was processed by the diamonds to an initial thickness of approximately 70 μm, and then a center hole with a diameter of 180 μm was drilled as the sample chamber. The Fe sample was loaded into the sample chamber along with several ruby chips to determine the pressure using the standard ruby fluorescent technique. Pressure calibration was performed using T64000 Triple Raman Spectrometer (HORIBA) at a wavelength of 532 nm. The experimental pressure ranged from atmospheric pressure to 26.9 GPa, with an excitation signal frequency of 173 Hz and a signal peak of −5 V to 5 V (peak-to-peak: 10 V). An excitation signal of lower frequency could inhibit the generation of eddy currents to a certain extent. All data were obtained under room temperature. In order to prevent any effect on the accuracy of the data during the experiment, no other high-power devices were present in the power grid that supplied the power for the system.

4. Results and discussion

The "stress–voltage" curve (Fig. 3) is plotted according to the recorded data. The magnetic properties of Fe under pressure and their associated structural phase transitions have been the subject of various theoretical studies. Fe is known to undergo a pressure-induced phase transition from the ferromagnetic body-centered cubic (bcc) α-phase to the nonmagnetic hexagonal-close-packed (hcp) ε-phase.[37,38] At room temperature and with the increasing of pressure, Fe exhibits phase transition at a pressure of $\sim$ 13 GPa.[10,16,39]

Figure 3 shows the plotted signal curves during the experiment. The DAC used in Fig. 3(a) is made of traditional magnetic hard alloy. An inflexion point is observed near 14 GPa. The large change in the background signal with the increasing of pressure completely covers the signal of the sample. The pressing machine used in Fig. 3(b) is fabricated using new materials. When the pressure is less than 10 GPa, the signal does not change with the increasing of pressure, which shows that the system overcomes the influence of the change in background noise composed of eddy current signals and magnetic signals because of the movement of DAC parts with the increase in pressure. As the pressure approaches to 13 GPa, signal change is observed. By measuring and comparing the inductance values of the two coils inside the detector it is shown that the magnetic permeability of the sample inside the sample chamber has been attenuated, which is indicated by the decline in the slop of the signal fitting line. With the further increase in pressure, the decrease in signal voltage becomes more evident. This observation suggests that in the transition of Fe from the bcc phase to hcp phase, the transformation from the ferromagnetic to the non-magnetic actually occurs. This finding is consistent with the previous theoretical prediction. However, given the existence of measurement deviations in the ruby–pressure calibration, the pressure to induce magnetic transition P_c could not be accurately calibrated. We could only determine that the magnetic transition and structural phase transition are in the same pressure region. Ensuring the simultaneous occurrence of the changes by plotting the curve with the increasing of pressure would provide significant evidence for supporting the existing theories.

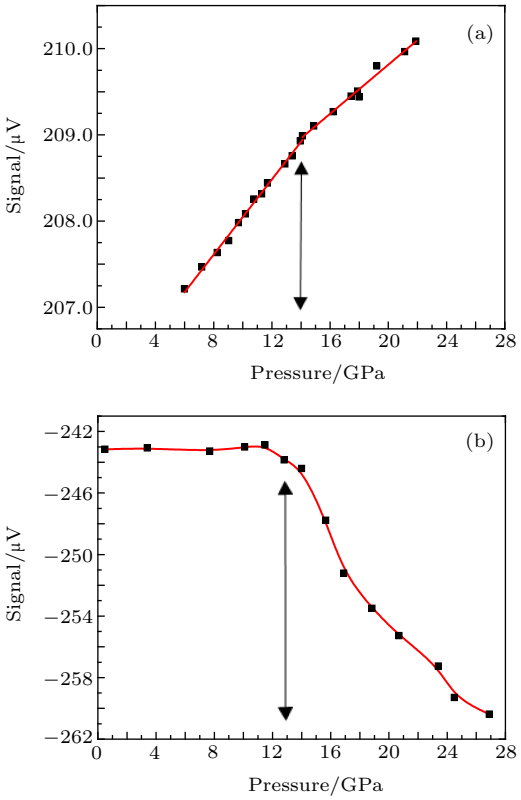


Fig. 3. (color online) Variation curves of magnetic permeability signals of the sample versus pressure, measured at room temperature. (a) Curve obtained by DAC made of traditional magnetic hard alloy. An inflexion is shown, but the signal for the sample is totally covered. (b) Curve obtained by DAC made from new materials. In the graph, the arrow points to the magnetic transformation site $P_c \approx 13$ GPa.

5. Conclusions

We measure the variation curves of the magnetic permeability signal of Fe and reach a pressure of 26.9 GPa using a new system. The signal shows an inflexion at the 13-GPa phase transition pressure. We confirm that the Fe sample undergoes a pressure-induced magnetic transition from the ferromagnetic phase to the nonmagnetic phase, verifying the accuracy of the previous theory. The system can be used to investigate the magnetic properties of materials under high pressure. This approach is also compatible with other existing measurement methods and has good development potential.

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