

In situ HPHT resistance measurement of $(\text{Fe}_{0.125}\text{Mg}_{0.875})_2\text{SiO}_4$ in a designed laser heated diamond anvil cell

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2007 J. Phys.: Condens. Matter 19 425210

(<http://iopscience.iop.org/0953-8984/19/42/425210>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 129.252.86.83

The article was downloaded on 29/05/2010 at 06:13

Please note that [terms and conditions apply](#).

In situ HPHT resistance measurement of $(\text{Fe}_{0.125}, \text{Mg}_{0.875})_2\text{SiO}_4$ in a designed laser heated diamond anvil cell

Ming Li^{1,2}, Chun Xiao Gao^{1,5}, Yan Zhang Ma³, Ai Min Hao¹,
Chun Yuan He¹, Xiao Wei Huang¹, Yan Chun Li⁴, Jing Liu⁴,
Hong Wu Liu¹ and Guang Tian Zou¹

¹ National Laboratory of Superhard Materials, Institute of Atomic and Molecular Physics, Jilin University, Changchun 130012, People's Republic of China

² Department of Physics and Chemistry, Henan Polytechnic University, Jiaozuo 454000, People's Republic of China

³ Department of Mechanical Engineering, Texas Tech University, Lubbock, TX 79409, USA

⁴ Beijing Synchrotron Radiation Laboratories, Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100039, People's Republic of China

E-mail: cxgao599@yahoo.com.cn

Received 3 August 2007

Published 18 September 2007

Online at stacks.iop.org/JPhysCM/19/425210

Abstract

A laser-heated diamond anvil cell was used in resistance measurements at high pressures and high temperatures. A thin film of alumina (Al_2O_3) was coated on both diamond anvils for thermal insulation before fabricating the microcircuit for resistance measurements. This technique significantly improved the heating conditions in the sample chamber. Using this technique, we measured the resistivity of $(\text{Fe}_{0.125}, \text{Mg}_{0.875})_2\text{SiO}_4$ to 35 GPa and 3450 K, which reveals an increase in the activation energy of the carrier with increasing pressure. The activation volume and activation energy are both less than the values at lower pressures and temperatures.

1. Introduction

Resistivity measurement at extreme conditions permits *in situ* investigations of the electron transportation behavior of matter such as pressure-induced superconductivity, insulator-to-metal transition, and magnetic susceptibility [1–5], and thus always attracts attention [6, 7]. Many techniques have been developed for resistance measurement under high pressure. For instance, Weir *et al* sputtered tungsten carbide film on a diamond anvil, which was then encapsulated by a layer of diamond for electrode protection [8]. This method provides

⁵ Author to whom any correspondence should be addressed.

protection to the microcircuit and significantly improves the technique of high-pressure resistance measurement. On the other hand, research results indicate that electronic and ionic transport properties are significantly different at high temperature from those at ambient temperature [6, 9–11]. It is therefore important to measure the resistivity *in situ* at high pressures and high temperatures (high P – T). However, most high-pressure resistance measurements have been carried out below about 1700 K [6, 10, 12] and [13]. *In situ* electrical conductivity measurement at many thousands of degrees, which is the most interesting temperature range for addressing scientific problems in material science and geophysics, has posed a challenge. Very recently, Gao *et al* sputtered and patterned molybdenum film on a diamond anvil facet to form electrodes [14, 15]. They covered the circuit with an alumina (Al_2O_3) film for electrical insulation, allowing accurate measurement of electrical conductivity at high pressure in a diamond anvil cell (DAC).

Moreover, a laser-heated DAC can generate thousands of degrees at pressures over a megabar [16–18]. Integration of the microcircuit with laser-heated diamond cell techniques would enable us to study the electronic transportation properties *in situ* at high P – T . Because of the high thermal conductivity of diamond, one important aspect of using a laser for sample heating in a DAC is sample insulation from the diamond anvils. In addition, as well as being used as electrical insulation material as described above, Al_2O_3 has also been used as a thermal insulation material [19]. The crystal structure of Al_2O_3 is stable to 175 GPa [20], which eliminates the error and complexity introduced in high-pressure resistivity measurement. Its large value of bulk modulus (253 GPa) can protect the electrical circuit from destruction during sample flow at high pressures [21]. Most importantly, Al_2O_3 is transparent in a large range of wavelengths, and thus permits both the introduction of a laser beam onto the sample for heating and the collection of black-body emission for temperature measurements. The high degree of hardness ($\sim 1500 \text{ kgf mm}^{-2}$), the non-conduction to very high pressures, as well as the high wearability are additional extraordinary properties that make Al_2O_3 an ideal material for both thermal and electrical insulation in high- P – T resistance measurement.

Materials that have $(\text{AB})_2\text{SiO}_4$ (olivine) structure can be found naturally and can be taken directly into industrial applications. Their properties dramatically change with high pressure and high temperature, thus they can be utilized for sensors under extremely high- P – T conditions. The chemical stability and structural phase transformation properties are extremely important for exploring and characterizing the interior of Earth and other planets. The intent of this paper is to develop the DAC for *in situ* resistance measurement at high P – T and apply it for some materials. Therefore, we fabricated an Al_2O_3 film on a diamond anvil for both electrical and thermal insulation in high- P – T resistivity measurement using a laser-heated diamond anvil cell. We thus measured the resistivity of a silicate to 35 GPa and 3450 K.

2. Experiments

In fabrication of the microcircuit on a diamond anvil for a laser-heated diamond anvil cell, we adopted the techniques developed by Gao *et al* [14, 15]. The difference is that our circuit constitution is based on the van der Pauw method, and only the thickness of the sample is needed. Moreover, the accuracy is improved noticeably. First, a $3 \mu\text{m}$ -thick alumina film was sputtered on the diamond anvil as a heat insulation layer. A molybdenum (Mo) film with a thickness of $0.3 \mu\text{m}$ was deposited on the alumina layer. The diamond was heated to 600 K to enhance the adhesion of the Mo film and the alumina layer in the deposition process. The Mo film was patterned into detecting electrodes according to the van der Pauw model using photolithography and chemical etching. Then, a layer of alumina with a thickness of $2 \mu\text{m}$ was sputtered on the molybdenum to encapsulate and protect the microcircuit. A detection

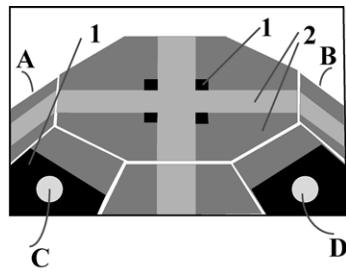


Figure 1. The completed designed microcircuit on a diamond anvil: 1 is the metal molybdenum; 2 is the alumina layer; A, B, C and D are the lead wires.

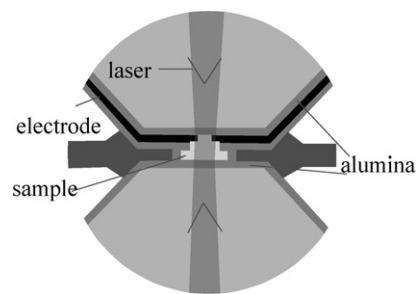


Figure 2. The cross-section of the designed DAC.

window was made to expose the electrode ends. Figure 1 shows the completed microcircuit. The distances between the opposite electrodes were $30\ \mu\text{m}$. This matched the area of uniform temperature distribution during laser heating. We used a Mao–Bell-type diamond cell in our experiment [22]. A T-301 steel gasket with preindentation was selected, with a hole of $150\ \mu\text{m}$ in diameter drilled in the center of the indentation. Fine Al_2O_3 powder was then filled and packed in the hole, into which a concentric hole $100\ \mu\text{m}$ in diameter was drilled, serving as a sample chamber. For further insulation of the electrodes and the gasket, both sides of the gasket were coated with a layer of Al_2O_3 ($2\ \mu\text{m}$ thick). The sample assembly in the diamond anvil cell for high- P - T resistivity measurement is shown in figure 2.

The atomic ratio of Mg/Fe in the sample measured in our experiment was determined by x-ray photon electron spectroscopy to be 0.875/0.125. Under the provided conditions, the sample falls into a perovskite structure phase $[(\text{Fe}, \text{Mg})\text{SiO}_3]$ and oxide $[(\text{Fe}, \text{Mg})\text{O}]$. The sample was packed in the sample chamber without a pressure medium. The double-sided laser-heating technique was used in the experiment [23–25]. The laser that was used was a Nd:YLF laser with a wavelength of $1053\ \text{nm}$ operated in the TEM_{01} transverse electric mode. The laser beam was evenly split and focused from both sides on the sample. A thermoelectrically cooled CCD detector equipped with an Acton SpectraPro300i spectrograph was used for the black-body spectrum measurement to determine the temperature of the sample. During the measurement, the laser power was kept at a pre-set level for 5 min before resistance measurement for the sample to reach thermal equilibrium. In the measurement, using the phase diagram of $(\text{Mg}, \text{Fe})_2\text{SiO}_4$ [26–28], we selected the particular P - T range in our experiment to avoid the complexity that may be caused by its transformation [29–31]. Figure 3 shows the temperature distribution on the heated sample at $3300\ \text{K}$. Within a $50\ \mu\text{m}$ range, the temperature varies

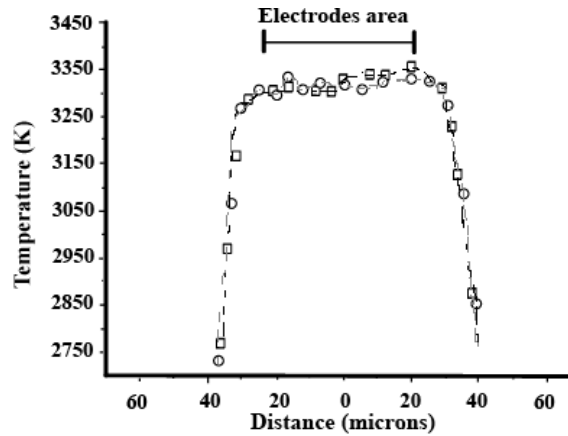


Figure 3. Temperature profile measured on both sides of the sample at 26 GPa.

within 50 K. The range of homogenous temperature is much larger than those reported by Shen *et al* and Ma *et al* [17, 32]. This demonstrates that the insulation conditions have been significantly improved in our experiments. This improvement substantially eliminated the error in resistance caused by the large temperature gradient, and thus greatly enhanced the accuracy of the measurements.

3. Results and discussion

In the resistance measurement, a $10 \mu\text{A}$ current (I_1) was introduced through electrode A (+) and B (–) while the voltage (V_1) between C (+) and D (–) was being recorded. The electrodes were then switched. A $10 \mu\text{A}$ current (I_2) was introduced again through A (+) and C (–) while the voltage (V_2) between B (+) and D (–) was being measured. This procedure eliminates errors induced by the electrodes [31]. For the calculation of resistivity, we measured the thickness of the sample at all pressures using a micrometer with a precision of $0.5 \mu\text{m}$ through measurement of the distance between the bottom facets of the anvils and subtraction of the thickness of the diamond anvils.

The electrical resistivity was determined from the van der Pauw equation [33]:

$$\exp(-\pi L R_A \sigma) + \exp(-\pi L R_B \sigma) = 1 \quad (1)$$

where $R_A = V_1/I_1$, $R_B = V_2/I_2$, L and σ are, respectively, the resistivities of electrodes A and B and the thickness and the electrical conductivity of the sample being tested. Figure 4 shows the electrical conductivity of $(\text{Mg, Fe})_2\text{SiO}_4$ under high P – T (under these experimental conditions, the olivine has decomposed to perovskite and oxide), and no melt is found from seeing the energy dispersive x-ray diffraction (EDXD) spectrum, which is consistent with the conclusion of Sweeney and Heinz [34]. It is found that the conductivity increases with increasing temperature but decreases with increasing pressure, and our experimental results are consistent with previous resistance measurements at lower temperatures [6, 35, 36]. Many silicate minerals are insulators, but they display the properties of an impure semiconductor under high P – T . It was proved that, at temperatures above 1200°C , both electrons and ions serve as the charge carriers for olivine material. Yet above 1390°C , the vacancy generated by Mg in olivine becomes the dominant charge carrier [10]. Our experimental result indicates that the ionic conduction mechanism plays the primary role in a much larger P – T range (35 GPa

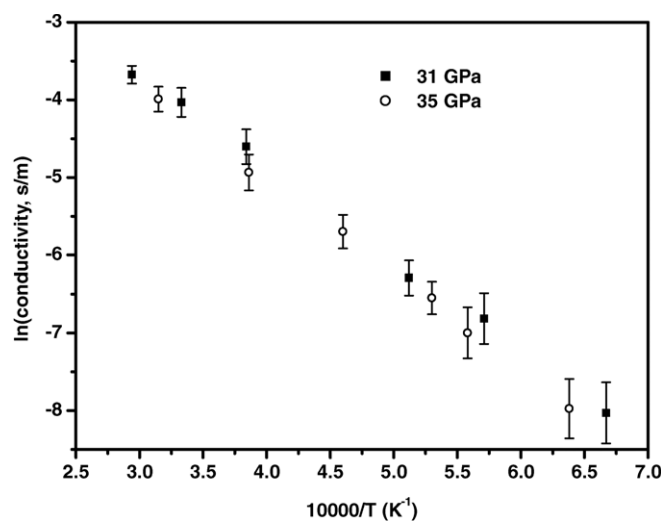


Figure 4. Electrical conductivity of the sample as a function of reciprocal temperature. Squares and circles denote the logarithm of the sample conductivity at 31 and 35 GPa.

and 3450 K). Considering electronic conduction being the conduction mechanism for olivine at lower temperatures (20–400 °C) [11], our experiment further demonstrated that the conduction mechanism for silicate differs at high temperature from that at low temperature.

With our experimental data, the activation enthalpy for conduction of the sample, ΔH , can be derived from $\sigma = \sigma_0 \exp(-\Delta H/K_B T)$, with σ and σ_0 being, respectively, the conductivity at the absolute temperatures of T and infinity, and K_B being the Boltzmann constant. The activation energy, ΔE , and the activation volume, ΔV , can be determined from $\Delta H = \Delta E + P\Delta V$, with P being the applied pressure [6, 11]. The activation energy is 0.926 and 0.952 eV when the pressure is 31 and 35 GPa, and the activation volume is $0.236 \text{ cm}^3 \text{ mol}^{-1}$. The value of activation energy increases with increasing pressure, but the effect is feeble. $\Delta V (= \partial \Delta H / \partial P)$ expresses the effect of the pressure on the energy barrier to charge transport, and we could find a general tendency for ΔV from the results of Xu *et al* and the present experiment, even though the starting sample is different [6]. The activation volume of our sample is much smaller than that of Xu *et al* in a $\text{Mg}_{1.8}\text{Fe}_{0.2}\text{SiO}_4$ sample at 4–10 GPa, indicating less lattice deformation in the present sample. The reason is the different ratio of Fe/Mg, as well as maybe the different phase (in our experiment, the silicate is γ phase with perovskite structure mixed with oxide, and it is α phase in the experiment of Xu *et al*). It is also found that the activation energy increases with increasing pressure. No significant change in activation energy can be associated clearly with the transformation of the sample covered in our P – T range.

In our experiment, the shape of the microcircuit was a square with fixed edge lengths. It solidly adhered to the surface of the diamond anvil. Thus, the change in distance between the electrodes during the experiment can be neglected, and would not introduce a large enough error to affect the accuracy of the measurement. The accuracy of the resistivity measurement is determined mainly by that of the thickness measurement in the experiment. In our experiment, the systematic error of the sample thickness measurement is less than $0.5 \mu\text{m}$. With the thickness of the sample being larger than $10 \mu\text{m}$ in the pressure range that we reached, the error in the thickness is less than 5%. Thus the maximum systematic error in our resistivity measurement is below 10% (table 1).

Table 1. The total error in the electrical conductivity of the sample.

31 GPa			35 GPa		
T (K)	$ \Delta\sigma $ (s m ⁻¹)	$ \Delta\sigma /\sigma$ (%)	T (K)	$ \Delta\sigma $ (s m ⁻¹)	$ \Delta\sigma /\sigma$ (%)
3450	0.049 48	6.2	3200	0.070 2	8.1
3000	0.080 57	9.2	2600	0.100 75	9.4
2600	0.096 98	9.7	2200	0.094 04	7.6
1950	0.097 02	7.1	1900	0.091	6.4
1750	0.142 03	9.6	1800	0.142 84	9.4
1500	0.170 88	9.8	1550	0.166 25	9.6

4. Conclusion

In summary, a technique has been developed for resistance measurement in an extended P – T range in a laser-heated diamond anvil cell. Al_2O_3 was utilized as both thermal and electric insulation in a laser-heated diamond anvil cell for resistance measurement, which significantly improved the heating conditions inside the sample chamber. This technique has many advantages, including efficiency in thermal and electrical insulation, high stability of the microelectronic circuit, simplicity in fabrication, and the capability for easy cleaning and reutilization of the diamond anvils. The conductivity of silicate has been measured to 35 GPa and 3450 K, with its high- P – T electrical properties being revealed.

Acknowledgments

This work was supported financially by the National Natural Science Foundation of China (grant nos 40473034, 40404007, 10574055 and 50532020) and the National Basic Research Program of China (grant no. 2005CB724404).

References

- [1] Eremets M I, Shimizu K, Kobayashi T C and Amaya K 1998 *J. Phys.: Condens. Matter* **10** 11519
- [2] Eremets M I, Gregoryanz E A, Struzhkin V V, Mao H-K, Hemley R J, Mulders N and Zimmerman N M 2000 *Phys. Rev. Lett.* **85** 2797
- [3] Ishizuka M, Amaya K and Endo S 1995 *Rev. Sci. Instrum.* **66** 3307
- [4] Struzhkin V, Hemley R J, Mao H-K and Timofeev Y A 1997 *Nature* **390** 382
- [5] Jackson D D, Aracne-Ruddle C, Malba V, Weir S T, Catledge S A and Vohra Y K 2003 *Rev. Sci. Instrum.* **74** 2467
- [6] Xu Y, Poe B T, Shankland T J and Rubie D C 1998 *Science* **280** 1415
- [7] Schock R N, Duba A G, Heard H C and Stromberg H D 1977 *High Pressure Research, Applications in Geophysics* ed M H Manghnani and S Akimoto (San Diego, CA: Academic) pp 39–51
- [8] Weir S T, Akella J, Ruddle C A, Vohra Y K and Catledge S A 2000 *Appl. Phys. Lett.* **77** 3400
- [9] Peyronneau J and Poirier J P 1989 *Nature* **342** 537
- [10] Schock R N, Duba A G and Shankland T J 1989 *J. Geophys. Res.* **94** 5829
- [11] Shankland T J, Peyronneau J and Poirier J P 1993 *Nature* **366** 453
- [12] Sakaoto D, Yoshiasa A, Yamanaka T, Ohtaka O and Ota K 2002 *J. Phys.: Condens. Matter* **14** 11375
- [13] Sakai N, Kajiwaru T, Tsuji K and Minomura S 1982 *Rev. Sci. Instrum.* **53** 499
- [14] Gao C *et al* 2005 *Rev. Sci. Instrum.* **76** 083912
- [15] Han Y *et al* 2005 *Appl. Phys. Lett.* **86** 064104
- [16] Heinz D L, Sweeney J S and Miller P 1991 *Rev. Sci. Instrum.* **62** 1586
- [17] Shen G, Rivers M L, Wang Y and Sutton S R 2001 *Rev. Sci. Instrum.* **72** 1273
- [18] Hu J, Xu J, Somayazulu M, Guo Q, Hemley R and Mao H K 2002 *J. Phys.: Condens. Matter* **14** 10479
- [19] Wirhford M J, Brown D J W and Piper J A 1996 *J. Phys. D: Appl. Phys.* **29** 315

- [20] Jephcoat A P, Hemley R J and Mao H K 1988 *Physica B* **150** 115
- [21] Richet R, Xu J and Mao H K 1988 *Phys. Chem. Miner.* **16** 207
- [22] Mao H K and Bell T M 1980 *Carnegie Institution of Washington Year Book* **79** 490
- [23] Shen G, Mao H K, Hemley R J, Duffy T S and Rivers M L 1998 *Geophys. Res. Lett.* **25** 373–6
- [24] Mao H K and Hemley R J 1998 *Ultrahigh Pressure Mineralogy: Physics and Chemistry of the Earth's Deep Interior*, *Rev. Min.* vol 37, ed R J Hemley (Washington DC: Mineralogical Society of America) pp 1–32
- [25] Zhou Q, Ma Y Z, Cui Q, Cui T, Zhang J, Xie Y, Yang K and Zou G 2004 *Rev. Sci. Instrum.* **75** 2432
- [26] Mao H K *et al* 1982 Iron–magnesium fractionation medal for the earth *High-Pressure Research in Geophysics* ed S I Akimoto and M H Manghnani (Tokyo: Center for Academic) pp 319–28
- [27] Knittle E and Jeanloz R 1987 *Science* **235** 668
- [28] Saxena S K *et al* 1996 *Science* **274** 1357
- [29] Liu L G 1975 *Geophys. Res. Lett.* **2** 417
- [30] Ito E and Takahashi E 1989 *J. Geophys. Res.* **94** 10637
- [31] Ito E, Akaogi M, Topor L and Navrotsky A 1990 *Science* **249** 1275
- [32] Ma Y Z, Somayazulu M, Shen G, Mao H K, Shu J F and Hemley R J 2004 *Phys. Earth Planet. Inter.* **143/144C** 455
- [33] van der Pauw L J 1958 *Philips Res. Rep.* **13** 1
- [34] Sweeney J S and Heinz D L 1998 *Geophys. Monogr. Ser. 101* (Washington DC: Am. Geophys. Union) p 197
- [35] Duba A, Heard H C and Schock R N 1974 *J. Geophys. Res.* **79** 1667
- [36] Constable S 1993 *J. Geomagn. Geoelectr.* **45** 707