

Magnetic-field-induced insulator-metal transition in $V_{1-x}W_xO_2$ ($x = 0.06, 0.12$) in magnetic fields of up to 500 T

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Abstract. The low-temperature insulator phase is found to be completely suppressed by ultrahigh magnetic fields of 500 T and transformed to the metallic phase in a $V_{1-x}W_xO_2$ ($x = 0.06$) thin film, which has a slightly lower metal-insulator transition temperature (T_{MI}) than that of the film reported in the previous ultrahigh magnetic field experiment [1,2]. It is also found that the insulator phase is more robust against a magnetic field for a highly W-doped film ($x = 0.12$), suggesting a different origin of the insulator phases between $x = 0.06$ and 0.12.

1. Introduction

Magnetic field effects on chemical bonding are an intriguing research subject; unique crystal structures and shapes of molecules, as well as resulting characteristic electronic states, are expected. It is known that any chemical bonding is impossible to create at a magnetic field greater than 10^8 T, which is realized on a neutron star. This phenomenon is termed chemical catastrophe (CC) [1,2].

A 1000 T class magnetic field is the greatest magnetic field available for condensed matter physics. The success of generating 1200 T using electromagnetic flux compression [3] has paved the way to exploring a new research area: ultrahigh magnetic field science.

Formation of multimer in solids, such as dimer or trimer, is in many cases a result of interplay between charge, orbital, and lattice degrees of freedom of electrons, and leads to intriguing physical phenomena such as the metal-insulator transition [4]. The multimer can be regarded as a molecule-like state, and the breaking of the multimer through the collapse of the molecular orbital by an ultrahigh magnetic field can be regarded as the solid-state version of the chemical catastrophe (SSV-CC). The SSV-CC has been discovered in W-doped VO_2 thin films at ultrahigh magnetic fields of up to 500 T [1,2]. In the present work, we have further investigated the SSV-CC in W-doped VO_2 using different thin films from the previous works.

2. Experimental methods

Two $V_{1-x}W_xO_2$ ($x = 0.06, 0.12$) thin films were grown on the TiO_2 substrate (0.5 mm thickness) with the pulsed laser deposition [5] and used for the experiment. The thickness of



the $x = 0.06$ film is 16 nm, and it is 21 nm for $x = 0.12$. The electrical resistance (R) and optical transmission (Tr) of these films have been measured at different temperatures in a zero magnetic field. The PPMS (Quantum Design) and the spectrometer (JASCO V-570) were employed for R and Tr measurements, respectively.

The magneto-transmission experiment with a laser light of $1.977 \mu\text{m}$ has been conducted in magnetic fields of up to 500 T. An ultrahigh magnetic field is generated by the electromagnetic flux compression [3]. A fiber laser (AP-Tm, AdValue Photonics) and an HgCdTe detector (Kolmer Technologies) were used for the magneto-transmission measurement. The $V_{1-x}W_xO_2$ thin film is sandwiched between the two optical fibers, which are for the irradiation and detection of a laser light. The transmission signal is recorded by a digitizer with 500 MHz sampling rate.

3. Results and discussion

Figure 1 shows the R of the two $V_{1-x}W_xO_2$ thin films. The metal-to-insulator transition (MIT) with decreasing temperature is observed. When we take the temperatures where R shows an inflection, as the MIT temperature (T_{MIT}), T_{MIT-} and T_{MIT+} are deduced to be 57.9 and 78.1 K, respectively, for the $x = 0.06$ film. For $x = 0.12$, T_{MIT-} and T_{MIT+} are 75.9 and 79.1 K, respectively. $- (+)$ in the subscript indicates the temperature decreasing (increasing) process. The T_{MIT} is in good agreement with the T_{MIT} vs W -content diagram reported previously [6].

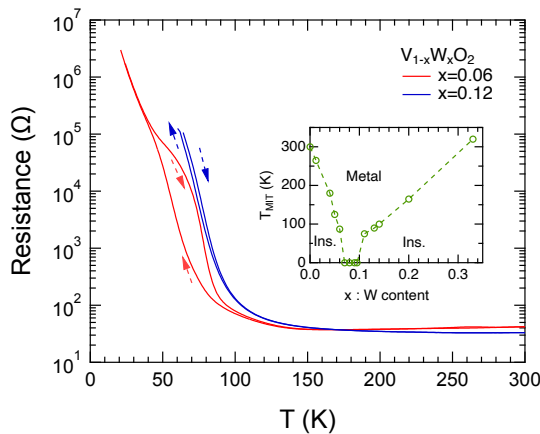


Fig. 1. The electrical resistance (R) is plotted as a function of the temperature. The schematic phase diagram, obtained from the results by Shibuya *et al.* [6], is shown in the inset. The ground state is insulating for $0 < x < 0.07$ and $0.1 < x < 0.33$.

The intriguing reentrant insulator-metal-insulator phase transition was reported with increasing W -content (x) [6]. T_{MIT} first decreases with x toward 0.07, and the metallic ground state appears ($T_{MIT} \sim 0$ K) when x is between 0.07 and 0.1. Then, an insulator phase appears when $x > 0.1$, and T_{MIT} increases with x up to approximately 0.33. The schematic phase diagram is shown in the inset of Fig. 1. Two insulator phases with lower and higher x are expected to postulate different origins for insulating [6].

In Figs. 2(a,b), the Tr in $x = 0.06$ and $x = 0.12$ films, respectively, are shown. The significant increase in the transmission at low temperatures reflects opening the energy gap in the insulator phase [1]. The vertical dashed line denotes the position at 0.627 eV that corresponds to the photon energy of $1.977 \mu\text{m}$ laser wavelength for the ultrahigh magnetic field experiment.

When we take the R at each temperature at which the Tr is measured, the relation between the R and Tr at $1.977\ \mu\text{m}$ is obtained. The R at different Tr is plotted in Fig.3(a,b). Although the Tr does not exhibit a monotonic temperature dependence as shown in Fig. 2(a,b), it is found to be possible to estimate the change in R by a change in Tr if the R shows at least a change larger than one order of magnitude.

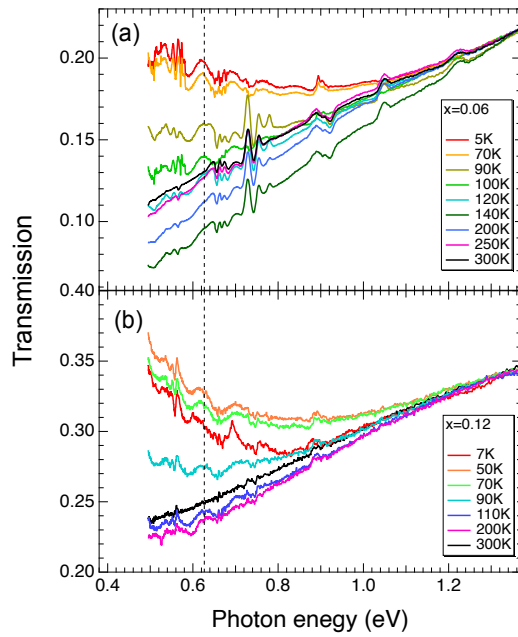


Fig. 2. (a) Optical transmission (Tr) at different temperatures in the $x = 0.06$ film. (b) Tr at different temperatures in the $x = 0.12$ film.

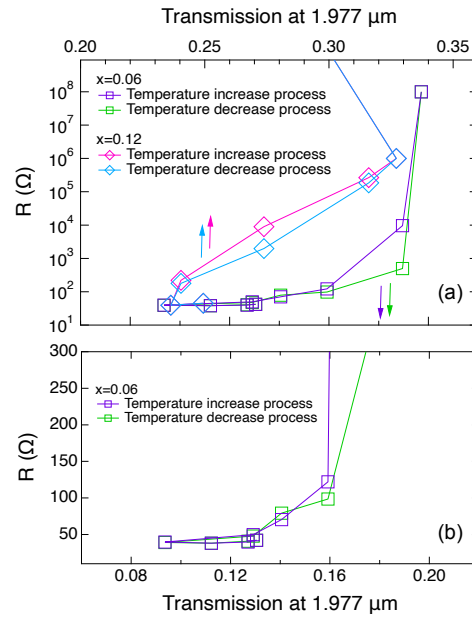


Fig. 3. (a) The electrical resistance R is plotted as a function of the optical transmission (Tr) at $1.977\ \mu\text{m}$ in $x = 0.06$ and 0.12 films. (b) The R is shown in more detail in the lower resistance region for the $x=0.06$ film.

The magneto-transmission in the $x=0.06$ film has been measured at $34\ \text{K}$, and the result is shown in Fig. 4 along with the magnetic field (B) as a function of time. The pickup coil for B detection and the $V_{1-x}W_xO_2$ ($x = 0.06$) film are found to have been destroyed at around $47.9\ \mu\text{s}$ when the magnetic-flux-compression process reaches the final stage and starts to implode. The data of the transmission and magnetic field after this timing has no physical meaning.

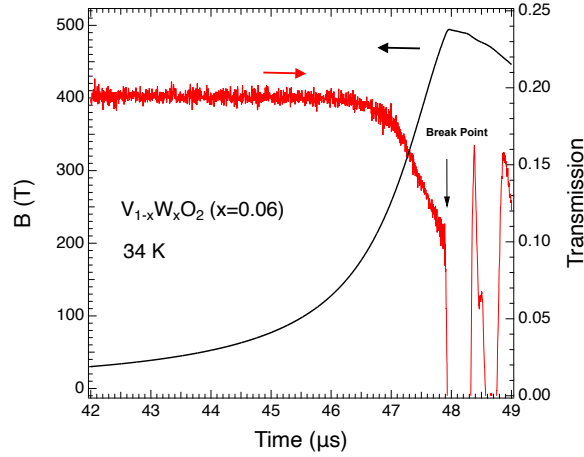


Fig. 4. The time variation of the optical transmission at 1.977 μm in $x = 0.06$, and that of a magnetic field (B).

A distinct decrease in the optical transmission starts to appear at around 46.5 μs , and reaches near 0.1 just before the destruction. This rather gradual decrease of the transmission is similar to the result of the previous study [1], and indicates the magnetic field-induced insulator-metal transition (IMT). According to Fig. 3, the change in the transmission from 0.2 to 0.1 corresponds to a 6 orders of magnitude decrease in R .

The optical transmission is replotted as a function of B in Fig. 5 (a). The two dashed lines indicate the transmission levels at 34 and 300 K at zero magnetic fields. If we define the transition magnetic field B_{IMT} by the field where the transmission passes the transmission level for the metallic phase at 300 K, B_{IMT} is found to be near 408 T. This value is smaller than $B_{\text{IMT}} \sim 500$ T in the previous study [1], where the different $\text{V}_{1-x}\text{W}_x\text{O}_2$ ($x = 0.06$) film (with higher T_{MIT} , i.e., $T_{\text{MIT-}}$ and $T_{\text{MIT+}}$, are 86.8 and 93.7 K, respectively) was measured. The origin of the different T_{MIT} between the two $x=0.06$ films is not very clear at present.

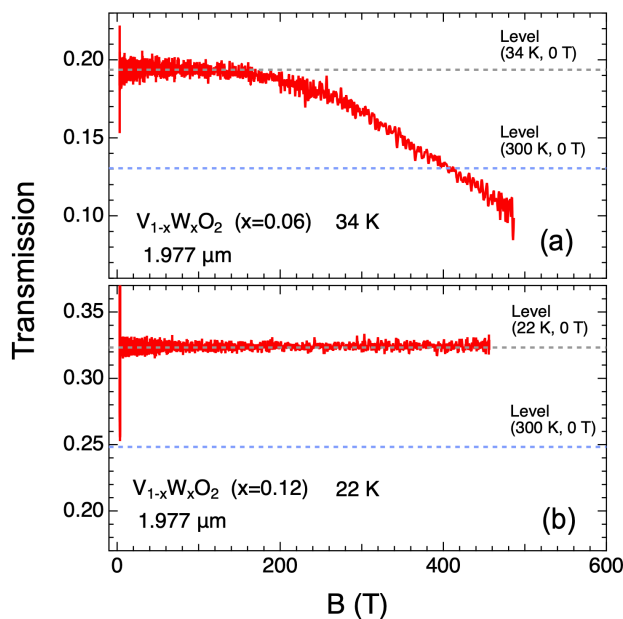


Fig. 5. (a) The magnetic field (B) variation of the optical transmission at 1.977 μm in $x = 0.06$. (b) The B variation of the optical transmission at 1.977 μm in $x = 0.12$.

The lower $B_{\text{IMT}} \sim 408$ T in the present work can be due to the lower T_{MIT} of the $\text{V}_{1-x}\text{W}_x\text{O}_2$ film, $T_{\text{MIT-}}$ and $T_{\text{MIT+}}$ are 57.9 and 78.1 K, respectively. In addition, the experimental

temperature 34 K in this work is higher than 14 K in the previous work, and this temperature difference may also be a reason for the lower B_{MIT} . Although further experiments are necessary to clarify the interplay between B_{MIT} and T_{MIT} , the observation of the field-induced IMT of another $x=0.06$ film in the multi-megagauss (several hundreds of Tesla) range has strengthened the validity of the occurrence of the SSV-CC in $\text{V}_{1-x}\text{W}_x\text{O}_2$.

A similar ultrahigh magnetic field experiment has also been conducted using the $x = 0.12$ film. The magneto-transmission is shown in Fig. 5 (b) in fields of up to 455 T. In contrast to the result of the $x=0.06$ film, the optical transmission shows no magnetic field dependence. As shown in Figs. 1 and 2, the MIT takes place in a similar temperature range as the $x = 0.06$ film. Therefore, the absence of the field-induced insulator-metal transition can be attributed to a different origin for the insulator phase at low temperatures in the $x = 0.12$ film.

It has been argued that the excess of electrons by the substitution of V with W decreases the valence of V from $4+$ to $3+$, and the formation of V-V dimers tends to be prohibited. The electron correlation for the $\text{V}^{3+}(d^2)$ possibly becomes the major origin of the insulating state of the high W-doping $\text{V}_{1-x}\text{W}_x\text{O}_2$ ($x > 0.09$) [6,7]. The significant difference between the results of the magneto-transmission in $x = 0.06$ and that of $x = 0.12$ supports this interpretation. In addition, one may come up with the question whether the field-induced metallic phase can appear at greater magnetic fields even in $x = 0.12$. If the $x=0.12$ film is a Mott insulator and can be understood within a simple single-band Hubbard model, the B -induced metallic phase would never happen because the electron correlation energy is generally at least one order of greater than the magnetic energy. In other words, the absence of the B effect on $x=0.12$ can give a hint to unveil the mechanism of the MIT at higher W-doped VO_2 . However, further experiments are required to answer this question, and are beyond the scope of this paper.

Finally, for $x = 0.06$, we speculate on the temperature dependence of the electrical resistance (R - T curve) in ultrahigh magnetic fields, assuming that the R - T curve just shifts to the lower temperatures by decreasing the T_{MIT} with a magnetic field. When we take the value of the optical transmission at 300 and 480 T, as shown in Fig. 5 (a), they are 0.16 and 0.11, respectively. Hence, using the relation shown in Fig. 3 (b), the corresponding R is estimated to be $120 \, \Omega$ at 300 T and $38 \, \Omega$ at 480 T. The speculated R - T curves are shown in Fig. 6 with dotted curves, which are given by just making a temperature shift of $\Delta T = 57$ and 113 K, respectively, so that $120 \, \Omega$ at 300 T and $38 \, \Omega$ at 480 T are obtained. Although the curve cannot be very accurate, this speculation can provide a potential R - T curve at an extremely high magnetic field. The ΔT corresponds to the shift of the T_{MIT} ; since 113 K is larger than the $T_{\text{MIT}} \sim 60 - 70$ K for the $x = 0.06$ film, a magnetic field of 480 T is expected to induce the metallic phase at the whole temperature range.

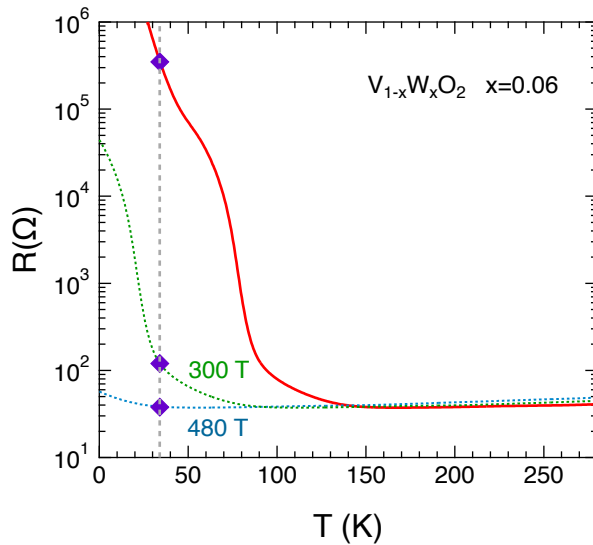


Fig. 6. The electrical resistance in the temperature increasing process at zero fields (red curve). The speculated temperature dependences at 300 and 480 T are shown with dotted green and blue curves, respectively. The vertical dashed line denotes the temperature (34 K) at which the ultrahigh magnetic field experiment has been done. The purple closed diamond represents the R at each magnetic field, which is evaluated from the magneto-transmission experiment.

4. Summary

The insulator-to-metal phase transition has been observed in a $V_{1-x}W_xO_2$ ($x = 0.06$) thin film, indicating collapse of the vanadium dimer through the suppression of the molecular orbital by an ultrahigh magnetic field of around 408 T. Regarding the $x = 0.12$ film, the insulator phase remains in magnetic fields of up to 455 T. The absence of the field-induced transition can be interpreted as indicating that the insulator phase originates from electron correlation, which is possibly rather insensitive to the magnetic field.

Acknowledgments

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