Discovery of the new superconductor MgB_2 and its recent development

by Yuji Zenitani and Jun Akimitsu

We recently discovered that the intermetallic compound magnesium diboride (MgB_2) exhibits the highest superconducting transition temperature (T_c = 39 K) of all metallic superconductors. In this paper we report on the basic superconducting characteristics of MgB_2 and the current status of the search for applications for this material. In particular, we summarize the research into its critical current (J_c) and critical magnetic field (H_c), which are important parameters for practical applications of this material.

1. Introduction

The search for practical applications of superconductors was spurred on by the discovery of high-temperature copper oxide superconductors in 1986, and after that every university research center and private corporation joined in the search for new superconductor materials and applications for them. But in practice, the difficulty of processing oxide-based superconductors prevented them from finding many practical applications. Although the superconducting transition temperature of oxide materials is substantially greater than that of metallic compound superconductors, finding practical applications for them still involved overcoming the hurdle of having to cool them to low temperatures. However, in the 15 or so years that have passed since then, remarkable progress has been made in the development of highly efficient cooling equipment that is able to cool superconductors without the use of liquid helium, and it now seems that practical applications for this equipment will soon begin to appear.

Although superconductors are thus faced with various obstacles on the path to practical applications, the reason why many people are looking for practical applications for these materials is that superconductors have the potential to radically alter the basis on which technology is built in the future. Specifically, it could lead to major developments such as: (i) a revolution in transport and distribution methods, such as linear motor cars, (ii) highly efficient electric power generation and lossless power distribution using superconductors, (iii) faster and higher-capacity information devices using superconducting integrated circuits, (iv) high-performance medical sensors such as MRI scanners, and applications to standard measurement devices, and (v) applications to small high-performance superconducting magnets, which would be useful for a wide range of industrial and commercial applications. Even in these broad categories, superconductors should find a wide range of applications by creating a technological paradigm shift.

But in order for superconductors to be widely applied until they achieve social and economic recognition, it is necessary to carefully ascertain the benefits and drawbacks of copper oxide high-temperature superconductors and intermetallic compound superconductors. We have investigated the possibility of superconductivity appearing in various different types of material from various different viewpoints, and recently discovered that magnesium diboride (MgB_2) has the highest superconducting transition temperature of all metal superconductors.

This paper describes how we made this discovery, and describes the superconductivity and other physical properties of MgB_2. We also report on our current expectations for future applications of this material.

2. MgB_2 : A New Superconductor

In intermetallic compound superconductors, which are referred to as BCS-based superconductors, or in superconductors that do not contain copper-oxide, the limit of the superconducting transition temperature has so far been in the region of 30 K, and no superconductors had been found to have higher superconducting transition temperatures. Until now, the highest superconducting transition temperature of an intermetallic compound superconductor was that of Nb_3Ge (23 K). However, we have discovered that the intermetallic compound MgB_2 exhibits superconductivity with a transition temperature of T_c=39 K, which is the highest yet recorded for an intermetallic compound.[1)]

3. Synthesis Method

Powdered magnesium (99.9% pure) and boron (amorphous, 99% pure) are mixed together in a molar ratio Mg:B = 1:2 inside a dry box filled with highly pure argon gas, and the mixture is press-formed into pellets. Pelletized samples are then wrapped in tantalum foil and placed in a graphite crucible, and using an O_2–Dr. HIP made by Kobe Steel Co. Ltd., they are fired at 973–1173 K for 1–10 hours in an atmosphere of argon gas at 196 MPa to obtain single-phase samples of MgB_2.

Other possible synthesis methods include sealed vacuum tube synthesis, ultra-high pressure pyrogenic synthesis, and synthesis by a solid-phase reaction in a stream of argon gas. However, the HIP method is got the most efficient results in MgB_2 samples with the greatest purity.

A possible reason for this is that magnesium has low melting and boiling points and reacts with elements such as oxygen to form highly stable compounds. When using an arc welding synthesis method, only the magnesium melts, and as a result the raw materials separate out into magnesium and boron. Ultra-high pressure pyrogenic synthesis entails very high costs for each time synthesis is performed. In the sealed vacuum tube synthesis method and the method involving low-temperature synthesis in a stream of argon gas,

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the magnesium can easily become oxidized.

When the inert gas argon is used in a hot hydrostatic compression method, the melting and boiling points of the magnesium are increased, and it becomes possible to synthesize MgB_2 at higher temperatures than in the above mentioned sealed vacuum tube synthesis method or the method involving low-temperature synthesis in a stream of argon gas. This method also suppresses the generation of impurities such as oxides and can synthesize single-phased MgB_2. It is also capable of synthesizing large volumes of samples at the same time. For these reasons, we used the O_2–Dr. HIP made by Kobe Steel Co. Ltd.

4. The Structure and Superconducting Characteristics of MgB_2

Figure 1 shows the X-ray diffraction spectrum of powdered MgB_2.

These results indicate that the material has a hexagonal crystal structure in space group *P6/mmm* (no. 191), where a = 0.3086 nm and c = 0.3524 nm. All the peaks have corresponding indices, so the sample must be single-phase. Figure 2 shows a model of MgB_2 crystal structure based on parameters derived from this structural analysis.

As you can see from this model, the structure is made from an alternating sequence of Mg and B_2 layers. The Mg layers consist of triangular lattice planes, and the B_2 layers consist of hexagonal honeycomb lattice planes similar to those of graphite. Since these planes have a two-dimensional structure, the material is expected to exhibit two-dimensional superconductivity as a result of its crystalline structure. The distance between adjacent B sites in the B_2 layers is about 0.178 nm, and the distance between Mg sites in the Mg layers is about 0.3086 nm, which is the same as the a-axis length. The Mg atoms are located on the central ax-

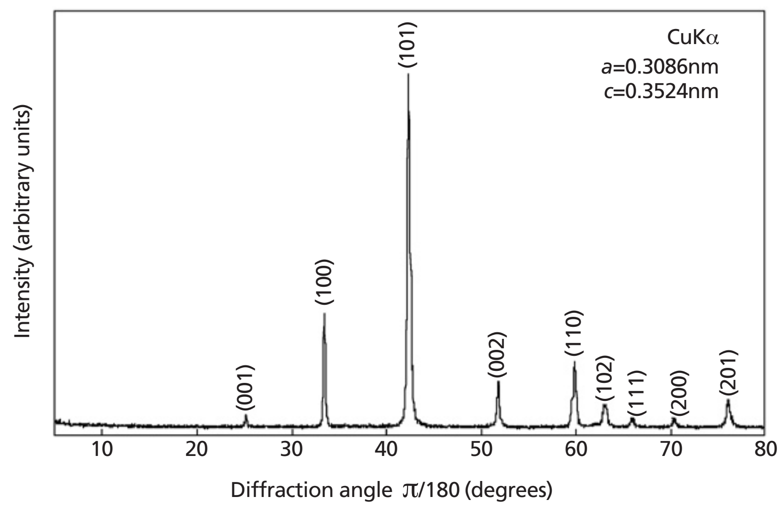

Fig. 1: X-ray diffraction spectrum of synthesized MgB_2. This spectrum corresponds to a hexagonal crystal structure in space group *P6/mmm* (no. 191), where a = 0.3086 nm and c = 0.3524 nm. All the peaks have corresponding indices, so the sample must be single-phase.

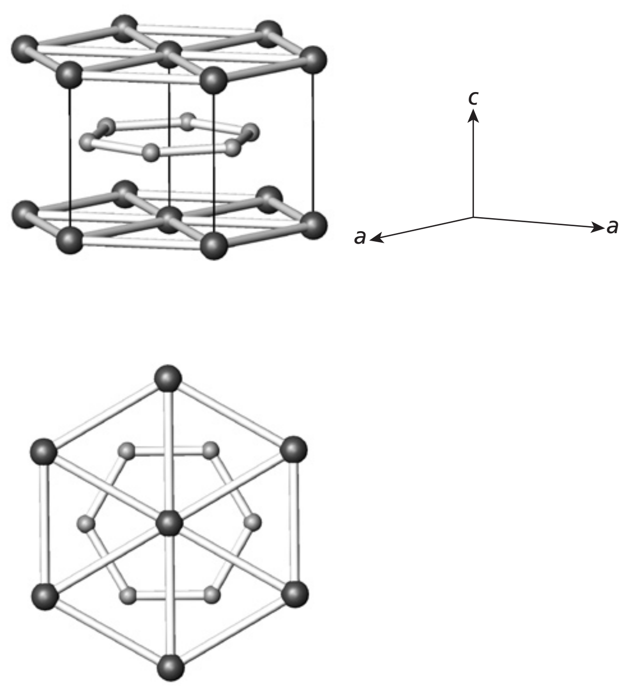

Fig. 2: The crystalline structure of MgB_2 based on the parameters derived from structural analysis. As the upper figure shows, the crystal consists of Mg planes containing only magnesium and B_2 planes containing only boron, which are layered alternately along the c axis. The lower figure shows the appearance from the [001] direction. The Mg layers consist of triangular lattice planes, and the B_2 layers consist of hexagonal honeycomb lattice planes similar to those of graphite.

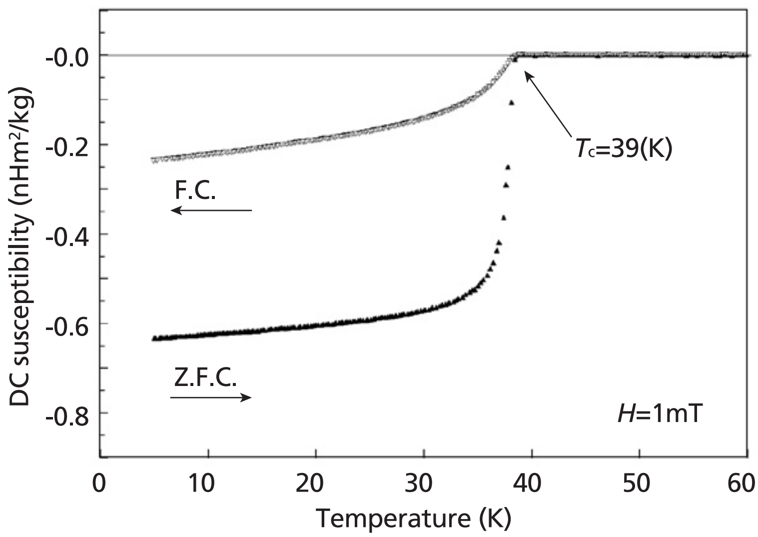

Fig. 3: The temperature variation of DC susceptibility in MgB_2. After the MgB_2 has been cooled to 5 K in a zero field, it is measured while increasing the temperature of the sample in an external field of 1 mT (zero field cooling: ZFC), and then its temperature is reduced again and measured in the same field (field cooling: FC). In both cooling processes it exhibits the Meissner paramagnetism characteristics of superconductors from 39 K, and the volumetric proportion of superconductor at 5 K as determined from the susceptibility when cooling in a magnetic field (FC) is found to be 49%.

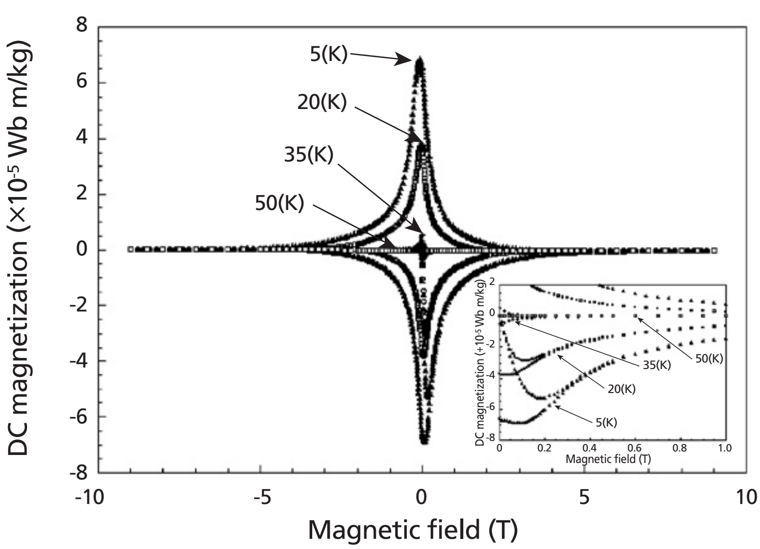

Fig. 4: Variation of DC magnetization with magnetic field in MgB_2. This graph shows the typical behavior seen in type 2 superconductors. Based on this graph, the estimated lower critical field H_{c1} and magnetic field penetration length at 0 K are approximately 75 mT and 97 nm, respectively.

is of the hexagonal columns formed by the B atoms. This structure is referred to as an AlB_2-type structure,[2,3] and is expressed by the general formula MB_2 (where M represents a metallic element).

Figure 3 shows how the DC susceptibility varies with temperature. After the sample has been cooled in the absence of a magnetic field, it is measured while increasing the temperature of the sample in an externally applied field of 1 mT (zero field cooling: ZFC), and then its temperature is reduced again and measured in the same field (field cooling: FC). As you can see from the results, it exhibits a large Meissner diamagnetism (a characteristic of superconductors) from 39 K in all the cooling processes, and the superconducting volume fraction at 5 K as determined from the susceptibility when cooling in a magnetic field (FC) is found to be 49%.

It can thus be seen that the superconducting transition temperature of MgB_2 is 39 K. Since the volumetric proportion of superconductor at 5 K as determined from the susceptibility in a magnetic field (FC) is 49%, we know that it is a bulk superconductor. Next, Fig. 4 shows how the DC magnetization depends on the magnetic field (*M-H* curve).

In the measurements made below the superconducting transition temperature, it exhibits typical behavior seen in type 2 superconductors, and based on this graph, the estimated lower critical field H_{c1} and magnetic field penetration length at 0 K are approximately 75 mT and 97 nm, respectively. The results of measurements made by other groups are reported as 28–48 mT and 85–180 nm. In general, when estimating H_{c1} from the magnetization curves of type 2 superconductors, any irregularities such as crystalline defects and impurities in the crystal act so as to pin the magnetic flux, so that the observed magnetization increases even when H_{c1} is exceeded, and it is thus important to take care when entering such discussions. This is probably the reason for the wide distribution in the values measured by each group.

Also, the magnetization of mixed states in a type 2 superconductor reflects the magnetic characteristics due to flux pinning, and it is possible to estimate the superconducting critical current J_c from the magnetization curves of flux in a trapped state using models such as those proposed by Bean[4] and Kim.[5]

Numerous reports have already been made regarding the variation of J_c with magnetic field and temperature in bulk samples, powder samples, wires, tapes and thin films. The values obtained in bulk samples are $\approx 10^5$ A/m^2 at H=0 T, $\approx 10^8$ A/m^2 at H=6 T, and $\approx 10^6$ A/m^2 at H=10 T.[6] These values are all 1–2 orders of magnitude smaller than those obtained with metallic superconductors that are already in practical use (Nb_3Sn and Nb-Ti). In powder samples, a value of 3×10^{10} A/m^2 is obtained at $H \leq 1$ T, which is larger than the value obtained with Nb_3Sn or Nb-Ti. However, it decreases rapidly in the presence of an external magnetic field, and it has been reported to drop down to 10^6 A/m^2 in a strong magnetic field of H=7 T.[7,8] Also, the value of J_c in wire[9-13] and tape[14-19] samples is lower than the values obtained from bulk and powder samples, especially at low magnetic fields. However, the attenuation resulting from external magnetic fields is suppressed due to the shielding effects of the processed shape, so these samples has a high J_c of 10^9 A/m^2 at H=5 T.

In other papers, Suo et al. reported that in tape processing the value of J_c could be increased approximately tenfold by an annealing process,[20] Wang et al. reported that in Fe coated wire processing it is possible to obtain performance similar to that of prolonged sintering by performing repeated sintering,[10] Jin et al. reported that in the processing of wires alloyed with metals such as Ti, Ag, Cu and Mo the value of J_c increases almost independently of T_c,[9] and Soltanian et al. reported on the shielding effect of the Fe sheath with respect to external magnetic fields in the processing of Fe-coated tape.[5]

Kim et al.,[2] Eom et al.,[22] and Paranthaman et al.[23] have reported on thin film samples, and suggest that it has a performance better than the value of 10^{10} A/m^2 obtained with Nb_3Sn and Nb-Ti, albeit at low magnetic fields. However, at high magnetic fields it has a J_c smaller than that of Nb_3Sn and Nb-Ti in the same way as a powder sample. As a method for addressing these drawbacks, Eom et al. also reported that it is possible to obtain a large J_c of 10^8 A/m^2 even under high fields of H=14 T by incorporating oxygen or MgO into the sample.

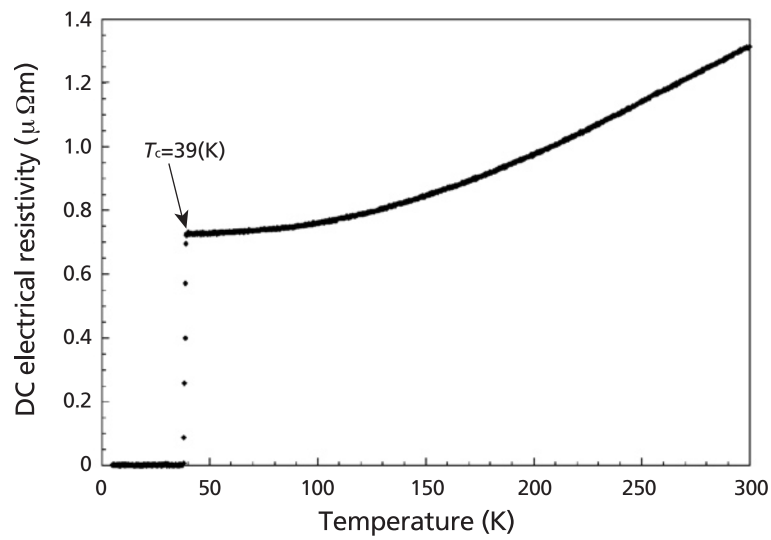

Fig. 5: The temperature dependence of electrical resistivity in MgB_2. The electrical resistivity decreases linearly from room temperature down to about 150 K, below which it decreases in proportion to the second or third power of temperature. Since a sintered sample was used for these measurements, it has a large electrical resistivity of approximately 0.7 μV/A·m above the superconductivity transition temperature (T_c). Like the results for the temperature dependence of DC susceptibility, the electrical resistivity drops sharply at 39 K, and at 38 K it exhibits zero resistance.

Figure 5 shows the temperature dependence of electrical resistivity. The electrical resistivity decreases linearly from room temperature down to about 150 K, below which it decreases in proportion to the second or third power of temperature.

For these measurements we used a sintered sample with a large grain size and a rather rough structure. As a result, the electrical resistivity above T_c is quite large at 0.7 μΩm. However, a much smaller value of approximately 0.0038 μΩm was obtained above T_c in measurements made with a dense sample obtained by ultra-high-pressure synthesis, and this is substantially lower than the value of 0.11 μΩm obtained with the conventional intermetallic compound superconductor Nb_3Sn. This low resistivity should allow it to transfer large electrical currents even in the normal conduction state.

The DC resistivity drops sharply at the same temperature of 39 K where a reduction in DC susceptibility is seen, becoming zero at 38 K. Even though the measured sample was not finely structured, a very narrow transition

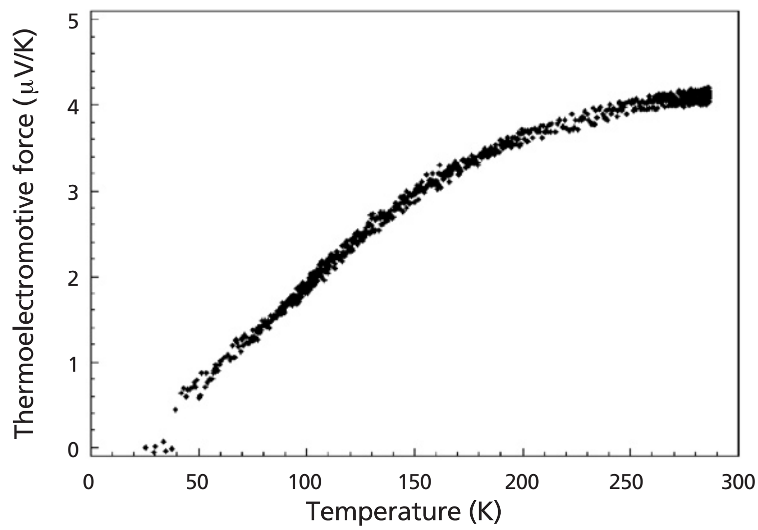

Fig. 6: The temperature dependence of thermoelectromotive force in MgB_2. After jumping discontinuously at the superconductivity transition temperature of 39 K, the sign of the thermoelectromotive force becomes positive (hole carriers) and increases in proportion to the temperature up to about 150 K (a behavior often exhibited by ordinary metals). As the temperature increases further, it approaches a fixed value.

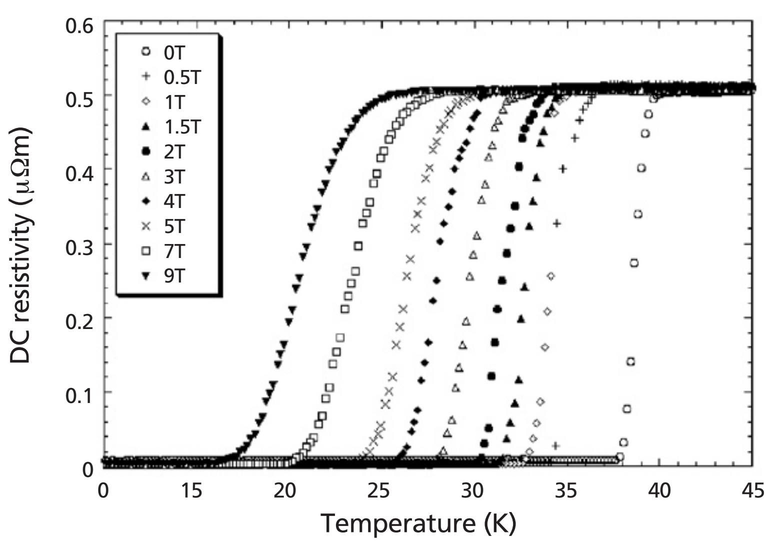

Fig. 7: Variation of electrical resistivity with temperature in MgB_2 in a magnetic field. As the applied magnetic field increases, the temperature at which the superconductivity transition starts and the temperature at which the resistance becomes zero both tend to decrease. This behavior is very similar to that observed in conventional metallic superconductors (e.g. A15 type superconductors).

range of about 1 K is still obtained, thus indicating the favorable nature of the superconducting characteristics.

Figure 6 shows the variation of thermoelectromotive force with temperature. After making a discontinuous jump at T_c, the sign of the thermoelectromotive force becomes positive (indicating that the carriers are holes), and increases in proportion to the temperature up to about 150 K (a behavior often exhibited by ordinary metals). As the temperature increases further, it approaches a fixed value.

This sort of temperature dependence variation is observed not only in the electrical resistivity as described above, but also in the Hall effect. Below 150 K, the proportional coefficient of the thermoelectromotive force is about 3–4 times the value for ordinary metals such as Cu and Au. However, it does not agree with the high carrier density value of 10^{23} cm^{-3} estimated from the Hall coefficient by other groups. Such a large carrier density is about 2 orders of magnitude greater than that of the intermetallic compound superconductor Nb_3Sn,[24] and one possible explanation for such a large figure is the existence of two types of carrier as mentioned above.

Next, Fig. 7 shows the temperature dependence of electrical resistivity in a magnetic field. As the applied magnetic field increases, the temperature at which the superconductivity transition starts and the temperature at which the resistance becomes zero both tend to decrease. This behavior is very similar to that observed in conventional metallic superconductors (e.g. A15 type superconductors), and contrasts with the behavior of copper oxide high-temperature superconductors (such as $YBa_2Cu_3O_7$).

Figure 8 shows the temperature variation of the upper critical magnetic field H_{c2} (T) as derived from the temperature dependence of electrical resistivity in a magnetic field.

At temperatures close to T_c, the temperature dependence of H_{c2} (T) is not proportional to temperature, and this behavior is also observed in intermetallic compound superconductors such as YNi_2B_2C. At other temperatures, its behavior is proportional to the temperature, and the gradient of this line is -0.69 T/K. Therefore, by considering this value together with the dirty limit in type 2 superconductors,[25,26] the value of H_{c2} (T) at 0 K is estimated to be approximately 18.6 T. Furthermore, the coherence length can be estimated to be about 4.18 nm from this value of H_{c2}, which basically agrees with the results of measurements made by other groups.

So far we have discussed the research into the basic superconducting characteristics of this material, but in addition many groups have reported on experimental and theoretical research into the mechanism whereby this superconductivity is expressed.

After our group reported the discovery of this superconductor, it was quickly followed by a report from Bud'ko et al. regarding experiments on isotopic effects.[27] Isotopic effects make it possible to ascertain whether or not the superconducting state is mediated by electron-phonon interactions as described by BCS (Bardeen−Cooper− Schrieffer) theory. According to their report, the onset of superconductivity occurs at 40.2 K in $Mg^{10}B_2$ and at 39.2 K in $Mg^{11}B_2$, and the coefficient α_B of the isotopic effect for B atoms is $\alpha_B = 0.26$, which is lower than the value of $\alpha_B = 0.5$ predicted by BCS theory. This result shows that this superconductivity is basically mediated by electron-phonon interactions, but this is not entirely so and it is expected that the superconducting state includes a complex mixture of other factors.

On the other hand, in experiments to determine the isotopic effects of ^{24}Mg and ^{26}Mg, Hinks et al. reported that there is almost no change in T_c and that the coefficient α_{Mg} of the isotopic effects of Mg atoms is about 0.02.[28] From these reports, it can easily be inferred that superconductivity in MgB_2 is strongly related to the mechanism whereby superconductivity is expressed in the B_2 layers.

Also, since there are numerous materials that have an AlB_2-type structure, it is easy to perform atomic substitution, and thus numerous attempts are being made to increase T_c particularly by carrier doping whereby the Mg sites (metal sites) are substituted with various other elements.[29] Substitution of the B sites with other elements is also being tried, and the effects of substituting them with carbon has already been reported.[30] However, nobody has yet reported any significant increases in T_c as a result of substitution with other elements. Although these experimental results suggest that it is difficult to increase T_c by substituting with other elements, the comparative ease with which elements can be substituted is worth investigating for application in other areas. However, it may be necessary to devise different approaches.

The superconducting gap formed in the superconducting state has been studied by a variety of spectral measurements, and is now broadly understood in terms of BCS theory. However, there are also many examples where different measurement techniques produce different results, so further clarification is still required. To address this situation, there is a need for detailed measurements using high purity single crystals.

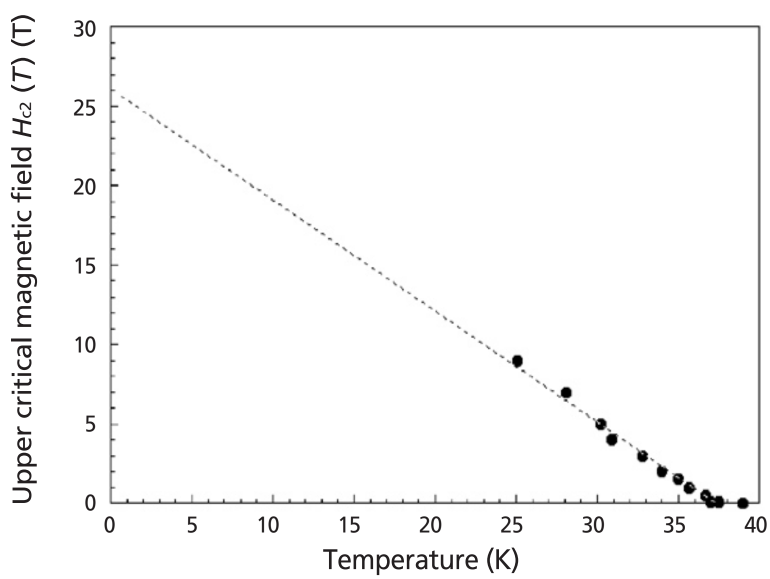

Fig. 8: The upper critical magnetic field H_{c2} of MgB_2 at various temperatures. This graph was obtained from the results of the electrical resistivity temperature dependence in a magnetic field by plotting the upper critical magnetic field H_{c2} at each temperature against the change in temperature. At temperatures close to the superconductivity transition temperature, the temperature dependence of H_{c2} is not proportional to temperature, and this behavior is also observed in intermetallic compound superconductors. At other temperatures, its behavior is proportional to the temperature.

The successful production of single crystals—albeit very small ones—was recently reported,[31)] and the anisotropic parameters obtained by making measurements such as those described above with single crystals have also been reported.

With regard to the H_{c2} anisotropy, a value of 2-3 has been reported for H_{c2ab}/H_{c2c}, and it has been suggested that this material has two-dimensional superconducting properties as expected from the crystalline structure, although this anisotropy is not as large as in oxide superconductors. However, in measurements made with single crystals, smaller values are estimated for H_{c2} and J_c than with other types of sample such as bulk samples, and it is thought that the higher values obtained for H_{c2} and J_c are due to extrinsic factors such as phase boundaries rather than intrinsic factors within the material.

5. The Development of Applications

As discussed above, although some of the most basic quantities of this material such as the superconducting transition temperature have been clarified, there is still a lack of reliable figures for the basic physical properties and characteristics of this material concerning other physical quantities of importance to practical applications, such as anisotropy. But even though the necessary characteristics are still unclear, MgB_2 is still regarded as a promising candidate for practical applications because it has several advantages such as the following:

(1)It has the highest superconducting transition temperature (39 K) of all intermetallic compounds.

(2)MgB_2 consists of the two elements magnesium and boron. It has fewer constituent elements than oxide superconductors, and since there is no uncertainty in its composition, it is a material that is easy to synthesize and stable.

(3)Compared with the intermetallic compound superconductors that have so far been used in practical applications, it consists of lighter atoms and thus weighs less.

First, (1) its superconducting transition temperature is about 20 K higher than that of conventional intermetallic superconductors. This has the advantage of providing a greater temperature margin for maintaining a superconducting state.

Next, (2) since it consists of just the two elements magnesium and boron, it can be made with fewer raw materials than copper oxide high-temperature superconductors and is comparatively easy to synthesize. And since MgB_2 has a stable composition and structure, its superconducting transition temperature does not vary greatly. In other words, it has the advantages of being a stable material that is easy to make. It also seems to lend itself to the production of forms such as thin films and wires.

Finally, (3) since it is a lightweight material that can even be used for power transmission lines, it can alleviate the load on power installations and the like because it functions as a superconductor.

Although this only a simple general view, MgB_2 has a number of advantages for practical applications.

Superconductivity, which is the most important attribute in terms of practical applications, is characterized by a critical current density (J_c) and an upper critical magnetic field (H_{c2}). In MgB_2, the upper critical magnetic field (H_{c2}) has been measured by a number of organizations, and its reported value for bulk materials is reported to lie in the range from H_{c2} (0) = 12 T to 18 T. The values obtained in measurements made with high-density bulk samples synthesized by ultra-high pressure synthesis range from H_{c2} (0) = 18 T to 26.5 T. The variation of these values arises because the quality of the samples is not constant. Compared with Nb_3Sn and Nb-Ti, which have already found practical applications, it has an intermediate value for H_{c2} (0), and the increase in dH_{c2}/dT is small. This indicates that it has a small mean free path.

Accordingly, it is predicted that it will be possible to improve H_{c2} by doping with other elements to reduce the mean free path. In practice, it has been reported that the value of H_{c2} in a thin film can be substantially increased by doping with oxygen. As for the critical current density (J_c), according to calculations using the Bean model from the history of magnetization curves, the value of J_c at 10 K and 10 kA/m is 5×10^9 A/m^2 inside grains and 0.4×10^9 between grains. The value of J_c at 5 K is lower than that of conventional superconductor materials such as Nb_3Sn, but it is thought that the J_c characteristics will improve as purer samples become available and the pinning mechanism becomes more clearly understood.

Today, research establishments all over the world are continuing to research the fundamental physical properties of MgB_2 and develop techniques for processing it into forms such as wires, tapes and thin films with a view to developing practical applications for this material. So far, although a large volume of data has been obtained by these establishments, it has not yet coalesced into an integrated model. But this is only to be expected from a material discovered so recently. For future study, comprehensive studies are required in areas such as the following:

(1)Why does MgB_2 have a high superconducting transition temperature of T_c = 39 K? To answer this question, the conformity of this material with conventional BCS theory needs to be studied, and a comparison with real experimental results is also required.

(2)The production of samples with higher purity and single-crystal samples. These samples could be used for more reliable and accurate measurements of the transport phenomena and magnetic/optical/thermal characteristics of MgB_2, thereby clarifying the mechanism for the expression of superconductivity.

(3)The establishment of processing techniques for the formation of wires, thin films and the like for practical applications, optimization of the processing parameters required for these techniques, and measurement of the superconducting characteristics of each form.

Finally, the reader should bear in mind that the authors of this paper are physicists–we are not the ideal people for finding practical applications for this material. As we write, a lot of research into MgB_2 is being conducted overseas, especially into its practical applications, and although this material was born in Japan it is in the process of being developed overseas. We would be much happier if this superconductor could be brought up in the country of its birth. We would like to finish by acknowledging the assistance received in this study by our research students J. Nagamatsu, N. Nakagawa, and T. Muranaka.

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