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Future prospects for NMR magnets: A perspective

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ABSTRACT

Superconducting magnet technology changed dramatically with the discovery of high temperature superconductors (HTS) in 1986, an event which drove the development of much higher field magnets. However, this technology paradigm shift has been delayed by as much as a decade in the case of NMR magnets. In this paper, we will provide a historical perspective to the reasons for this delay and assess the future prospects for high- and ultrahigh-field NMR magnets resulting from current trends in the development of HTS magnet technology.

1. Introduction

Since the discovery of high temperature superconductors (HTS) in 1986, superconducting magnet technology has changed beyond recognition. However, such a paradigm-shift has not appeared so far in NMR magnet technology. If we carefully examine the present status of HTS magnet technology, we can predict the future of NMR magnets over the next tens of years.

In this paper, we first provide a brief overview of NMR magnets and then predict the future of NMR magnet technology, with special attention to current HTS magnet technology.

2. Historical overview

When the NMR spectrometer was first developed in 1952, the available magnetic field was <100 MHz as iron-core electromagnets were used. A decade later, in 1964, saw the celebration of the first *superconducting NMR magnet* (200 MHz) developed by Varian Associates [1]. It was the beginning of the new era of high-field NMR magnets. Nine years prior this event, the first superconducting magnet with a field of 0.71 T was developed by Yntema et al. (1955) [2], using niobium (Nb) superconducting wire. Subsequently, "*niobium*" became a *magic word* for low-temperature superconductor (LTS) magnet technology. In 1961, "the International Conference on High Magnetic Fields" was held at MIT [3], demonstrating many high-field superconducting magnets, made by various kinds of promising superconducting wires, such as wires of an alloy of *niobium and zirconium* (NbZr) and wires of intermetallic compound of *niobium and tin* (Nb_3Sn). The excitement generated by this conference accelerated the development of the above described 200 MHz NMR magnet; this magnet was wound with NbZr wire, and included most of the necessary tools for NMR measurements, such as a shim coil system, a persistent-current circuit and a field stabilization system.

The NbZr wire was gradually replaced by a wire of alloy of *niobium and titanium* (NbTi), as it had the higher ductility necessary for wire drawing. The superconducting magnets at that time were frequently quenched due to flux jump [4]; this phenomenon was an abrupt magnetic flux motion during the coil charge, generating heat within the wire, resulting in coil quenches. The Rutherford High Energy Laboratory [5] demonstrated in 1970 that flux jump could be removed by using a multi-filamentary type of NbTi wire as shown in Fig. 1; numbers of NbTi fine filaments were embedded in copper- matrix, and twisted moderately. The upper critical field, H_{c2}, of NbTi is ~11 T at 4.2 K, hence it was possible to produce a 400 MHz (i.e. 9.4 T) NMR magnet by the end of the 1970s; H_{c2} is the magnetic field above which superconductivity disappears in type II superconductors. Both spatial homogeneity and temporal stability were further improved by using this type of wire, as it reduced the influence of the screening current (i.e. magnetization) induced in the wire during the coil charge.

For further enhancement of the NMR magnetic field, the use of a Nb_3Sn inner coil was necessary (i.e. a NbTi/Nb_3Sn coil), as the H_{c2}

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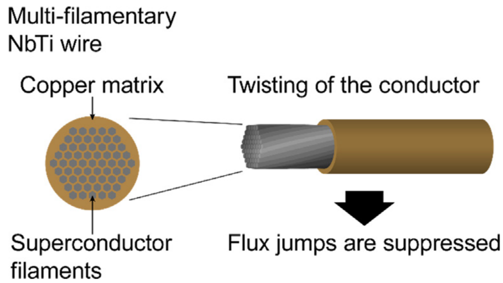

Fig. 1. Cross-sectional view of the multi-filamentary NbTi superconducting wire [5]; numbers of NbTi filaments, 50–100 μm in diameter, are embedded in the copper matrix. The wire is finally twisted to reduce inductive coupling.

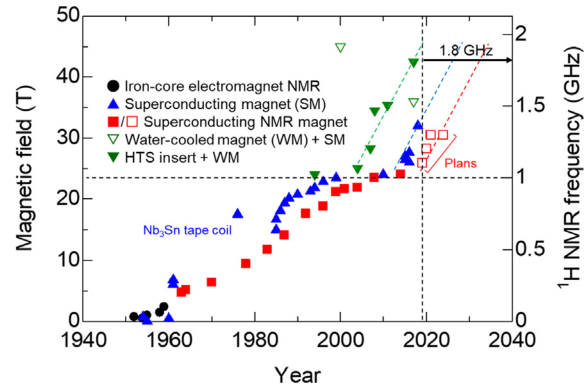

Fig. 2. History of the magnetic field strength for laboratory magnets and NMR magnets. After the discovery of the HTS in 1986, the magnetic field strength for laboratory magnets (▼, ▲) increased rapidly as a magnetic field resistant HTS inner coil was utilized; to the contrary, the magnetic field strength for NMR magnets (■) is still close to 1.0 GHz (i.e. 23.5 T) at present. The next generation of NMR magnets (□) are being developed; a persistent mode 1.3 GHz NMR magnet will be completed by 2024. From the view point of magnet technology, a 1.8 GHz (42.3 T) NMR magnet is possible, although there are obstacles regarding the use of REBCO wire.

for Nb_3Sn was ~20 T at 4.2 K. However, two challenges had to be overcome to accomplish this: the first challenge was to design a new fabrication process for the multi-filamentary Nb_3Sn wire. Thus far Nb_3Sn tapes had been used for a long period of time [4]; their drawbacks are a large influence of the screening current (i.e. magnetization) on the magnetic field and the frequent occurrence of flux jumps. In the 1970s, a new fabrication process called the "bronze process" was developed by Tachikawa et al. [6], enabling a multi-filamentary Nb_3Sn wire. The second challenge was developing a superconducting joint, as Nb_3Sn filaments were so brittle that a cold pressed joining process is impossible; an example of an indirect type of superconducting joint using superconducting solder is shown in Ref. [7]. NMR magnets accordingly reached 500 MHz (11.75 T) at the beginning of the 1980s.

Further improvements were also made by Tachikawa et al. [8], who demonstrated that the H_{c2} of the Nb_3Sn wire was enhanced if titanium (Ti) or tantalum (Ta) elements were added to the bronze-matrix or Nb filaments. Thus, 17 T class NbTi/Nb_3Sn magnets were made in the laboratories, resulting in production of a 750 MHz (17.6 T) NMR in the 1990s [9].

If an NMR coil is cooled by subcooled superfluid-helium at ~2 K, the magnetic field will be enhanced by 2–3 T due to the increase in H_{c2} [10] at ~2 K. A combination of doped Nb_3Sn and a 2 K cooling-system enhanced the magnetic field of the NMR magnet to 900 MHz (21.15 T) in the early 2000s. Bruker finally reached 1 GHz (23.5 T) in 2009 [11], which is still a world record of the magnetic field achieved using LTS NMR magnets.

Thus, the achieved magnetic field strength of NMR magnets has increased from 200 MHz to 1 GHz over the past 55 years as seen in Fig. 2, which was driven by the improvements in superconductors and liquid helium cooling systems. As shown in Fig. 2, the magnetic field strength for NMR magnets indicated by the red squares (■) followed that for the laboratory superconducting magnet shown by the blue triangles (▲) with a time delay of 8–10 years, as additional technologies are necessary for developing an NMR magnet.

3. State-of-the-art and future prospects

3.1. High temperature superconductors

Magnet technology dramatically changed in 1986 with the discovery of high temperature superconductors (HTS) by Bednorz and Müller [12], who were awarded the Nobel Prize in physics. A HTS is a superconductor with an unusually high critical-temperature (T_c) compared to that of a conventional low temperature superconductor (LTS) such as NbTi or Nb_3Sn; the T_c is a temperature above which superconductivity disappears. Three copper-oxide HTS wires such as $Bi_2Sr_2Ca_2Cu_3O_{10-\delta}$(Bi-2223, 1988 [13]) and $Bi_2Sr_2Ca_1Cu_2O_{8+\delta}$(Bi-2212, 1988 [14]) and RE (rare earth) $Ba_2Cu_3O_{7-\delta}$(REBCO, 1987 [15]) are the most promising for high-field magnets. The critical temperatures for LTS had been <23 K, whereas those for the above HTSs were in the range 85–110 K. As H_{c2} for HTSs is higher than 100 T [16], they are suitable for use as ultra-high field magnets. Thus, "*copper-oxide*" is a *magic word* for the HTS magnet technology

The Bi-2223 wire is a silver-matrix multi-filamentary tape wire as seen in Fig. 3(a), typically 4.5 mm in width and 0.3 mm in thickness. It is reinforced by metal plates bonded on both sides; the allowable tensile stress is 200–400 MPa. The Bi-2212 wire is a silver matrix multi-filamentary round wire, ~1 mm in diameter (Fig. 3(b)). The REBCO wire is a laminate structure tape wire (Fig. 3(c)). A buffer layer is produced on a strong metal substrate, and a thin REBCO layer is formed on it. It is typically 4 mm in width

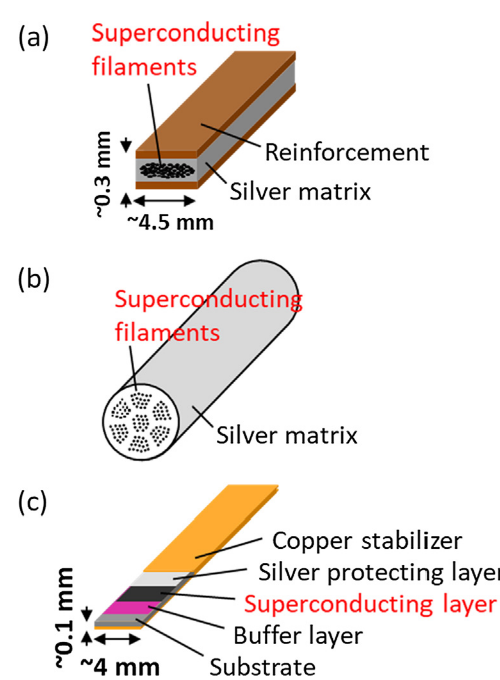

Fig. 3. Structures of the HTS wires: (a) A silver matrix multi-filamentary Bi-2223 tape wire, (b) A silver matrix multi-filamentary Bi-2212 round wire, and (c) A laminate structured REBCO tape wire.

and 0.1 mm in thickness and the allowable tensile stress is 500–700 MPa.

The critical current density (J_c) vs. magnetic field curves are compared in Fig. 4 for LTSs and HTSs [16–19]; the critical current density is the current density above which normal conductivity appears. As described above, J_c for NbTi [17] and Nb_3Sn [18] decreases steeply above 10 T and 23 T respectively; to the contrary, those for HTS wires do not notably decrease with the magnetic field in the range 20–30 T at 4.2 K. In Table 1, wire properties are compared; from the view point of magnetic field generation, REBCO is preferred to Bi-2223 and Bi-2212, while from the view point of field correction and field stabilization, Bi-2212 and Bi-2223 are preferred to REBCO. Therefore, choice of the HTS wire depends on the magnet design.

3.2. Generation of ultra-high magnetic fields

The first application of an HTS wire to a high-field magnet was made by Ohkura et al. in 1995 [20]; the magnet comprised a Bi-2223 insert and a backup-coil (Hybrid III at MIT). It generated 24 T under a backup field of 22.54 T, a world record at that time. Similar experiments using a water-cooled resistive magnet (WM) have been conducted thereafter; they were mostly conducted by the National High Magnetic Field Laboratory (NHMFL) in the USA. The magnetic fields thus achieved are indicated by the inverted green triangles (▼) in Fig. 2. In the case of a WM/Bi-2212 magnet, the magnetic field was <25 T [21] at first, later it reached 32.1 T in 2008 owing to improvements in the reaction process used to make the wires. Commercialization of the REBCO wire in 2009 changed this situation; as seen in Fig. 2, the magnetic field for WM/REBCO magnets attained 42.5 T lately [22], that is very

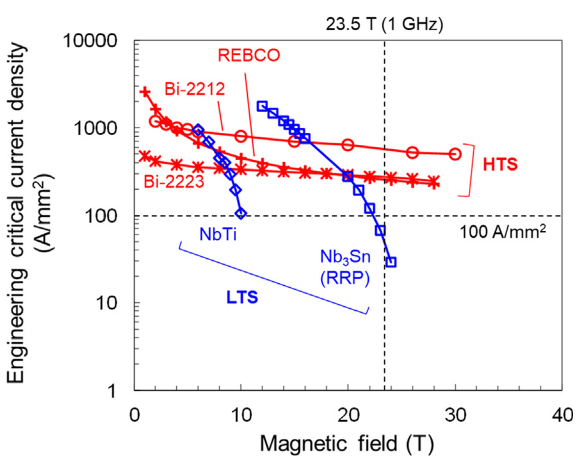

Fig. 4. Engineering critical current density vs. magnetic field for LTS wires such as NbTi [17] and Nb_3Sn [18] and HTS wires such as Bi-2223 [19], Bi2212 [16], and REBCO [19], measured at 4.2 K; the engineering current density is the critical current divided by the cross-sectional area of the wire. The critical current density decays rapidly above 10 T and 23 T for NbTi wire and Nb_3Sn wire, respectively. To the contrary, the critical current densities for HTS wires do not show notable decay in the magnetic field range 20–30 T. Figure adapted from [41].

close to the world record of the DC magnetic field of 45 T [23]; the 45 T magnet comprises a water-cooled inner resistive magnet and an LTS outer magnet (see Fig. 2). The magnetic field is close to an upper limit set by the experimental procedure, as the size of the HTS inner coil is limited by the bore diameter of the water-cooled resistive outer magnet, 40–50 mm.

Although a test of a WM/HTS magnet is the simplest way to demonstrate the feasibility of high field generation for HTS coils, a more straightforward and practical way is to manufacture an all-superconducting magnet such as an LTS/HTS magnet. The magnetic field achieved by such magnets is shown by the blue triangles (▲) in Fig. 2. A record-setting LTS/REBCO 32 T magnet was manufactured by the NHMFL in 2017 [24], while the runner-up is the 27.6 T LTS/Bi-2223/REBCO magnet made by RIKEN et al. [25]. Recently, the NHMFL launch a research and development effort for the next generation high field magnet such as a 40 T all-superconducting magnet [26], corresponding to 1.7 GHz in NMR frequency.

On the other hand, as seen by the red closed squares (■) in Fig. 2, the magnetic field for NMR magnets is still near 23.5 T (1 GHz) at present, although a Japanese team, including current authors, succeeded in developing a 1.02 GHz (24.0 T) LTS/Bi-2223 NMR magnet in 2014 [27]; it was driven by an external DC current supply (i.e. driven mode operation). Considering the extremely high potential of the HTS coil for generating a high magnetic field, it is obvious that the increment of the magnetic field is unsatisfactory. As seen in Fig. 2, there is a significant gap between the magnetic field strength for laboratory magnets (▼, ▲) and that for NMR magnets (■). It is due to the difficulty (a) in overcoming the harmful influence of the screening current induced magnetic field (i.e. magnetization effect) and (b) in developing superconducting joints between HTS wires; the screening current induced magnetic field is the magnetic field generated by the screening current that is induced during the coil charge [28].

Further projects in the development of ultra-high field NMR magnets are being advanced lately all over the world: (i) Bruker-BioSpin GmbH in the EU is currently developing persistent-mode 1.1 GHz (25.8 T) and 1.2 GHz (28.2 T) LTS/REBCO NMR magnets; it has not been revealed so far whether they will install superconducting joints between HTS wires or if they will use normal conducting solder-joints as described in their patent [29]. (ii) MIT in the USA is developing a 1.3 GHz (30.5 T) LTS/REBCO NMR magnet operated in the driven mode [30]. As the current density of the REBCO inner coil is extremely high due to the use of the no-insulation winding method, the magnet is nearly as small as a 700–800 MHz LTS NMR magnet. (iii) A Japanese team, including current authors, commenced a MIRAI Program in 2017 to develop a persistent-mode 1.3 GHz LTS/HTS NMR magnet [31]. According to the preliminary design, it comprises a REBCO innermost coil, Bi-2223 middle coils, and LTS outer coils. As the NMR magnet will be operated in persistent current mode, superconducting joints between HTS wires will be developed as described later. The magnet will be completed by 2024.

The magnetic field strength for these NMR magnets is shown in Fig. 2 by the red open squares (□). It is obvious that NMR magnets (red dashed line) follows the increase for the laboratory magnets

Table 1
Comparison of three promising HTS wires.

	Bi-2223 wire	Bi-2212 wire	REBCO wire
Critical current density at 30 T at 4.2 K	Lowest	Middle	Highest
Stress tolerance	200–400 MPa	200 MPa	500–700 MPa
Effect of screening current	Medium	Smallest	Largest
Single piece length of the HTS wire (2019)	<1 km	A few hundred m	<500 m

(green and blue dashed lines) with a time delay of 5–10 years. Considering the current upper limit of the magnetic field for laboratory magnets, i.e. 42.5 T, seen in Fig. 2, 1.8 GHz (42.3 T) may be a practical target for future NMR magnets.

An example of a preliminary design of a 1.8 GHz (42.3 T) LTS/REBCO NMR magnet is shown in Fig. 5. The size of the coil is assumed to be nearly the same as that for a 1.02 GHz LTS/HTS NMR magnet [27]; the red-colored zone shows the REBCO main coil, while the blue colored zone is an LTS correction coil. As seen in Fig. 5, the overall current density of the REBCO coil is 100–200 A/mm^2, while that of the LTS coil is ~100 A/mm^2; the circumferential tensile stress made by the electromagnetic force is <600 MPa. As the magnet is mostly made of high current density REBCO coils, it is extremely compact in size. However, the total length of the REBCO wire required is ~1400 km, which corresponds a wire costs of ~70 million US$.

Iwasa et al. [32] demonstrated a first-cut design of a 100 T magnet using REBCO wires; it is 20 mm in inner diameter, 5.6 m in outer diameter, and 16.7 m in coil height; i.e. it is an awfully big magnet. Based on his design, we may expect that an NMR magnet operated beyond 1.8 GHz may be achievable in the future. However, before such a development, we have to overcome obstacles regarding the REBCO wire, in particular the short lengths of REBCO wire currently available and its enormously high cost. Collaboration between the Applied Superconductivity society and NMR society will be necessary in this regard.

3.3. Persistent current mode operation

Although some preliminary studies of superconducting joints have been reported, those sufficient for NMR measurements have not been developed; a joint resistance of <10^{-12} Ω and a joint critical current of >300 A are required. In particular, a superconducting joint between REBCO wires has never succeeded so far. Park et al. [33] demonstrated the world's first REBCO lap joint in 2014, which was further improved by Ohki et al. [34] as an indirect type of superconducting joint. Indirect types of superconducting joints between Bi-2223 wires are also developed by Takeda et al. [35]. These joints will be installed in the persistent mode 1.3 GHz NMR magnet in the MIRAI Program [30]; based on NMR spectra, the properties of these superconducting joints and their applicability to the NMR magnet will be evaluated. A superconducting joint between Bi-2212 wires was developed by P. Chen et al. (NHMFL) in 2016 [36].

Fig. 6 shows the available working zone for superconducting joints; the horizontal axis shows temperature and the vertical axis is magnetic field. The available area of an LTS joint is limited to 2–9 K. On the contrary, the available area of HTS joints is as broad as 2–77 K in temperature, and 0–20 T in magnetic field. In fact, an excellent persistent current was proved for a small REBCO coil at 77 K [34].

3.4. Cooling systems

Frequent shortages and price hikes of liquid helium increase the need for a cryogen-free type of NMR magnet. Recent advances in

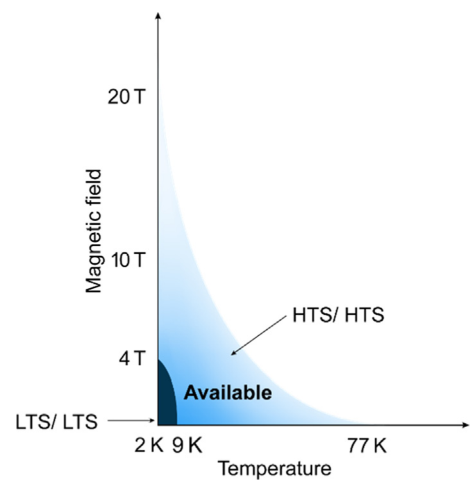

Fig. 6. Available region of superconducting joints; horizontal axis is temperature, and vertical axis magnetic field. For an LTS/LTS joint, the available area is limited to a small area near 4 K, while that for an HTS/HTS joint is much broader; the available range is 2–77 K in temperature and 0–20 T in magnetic field.

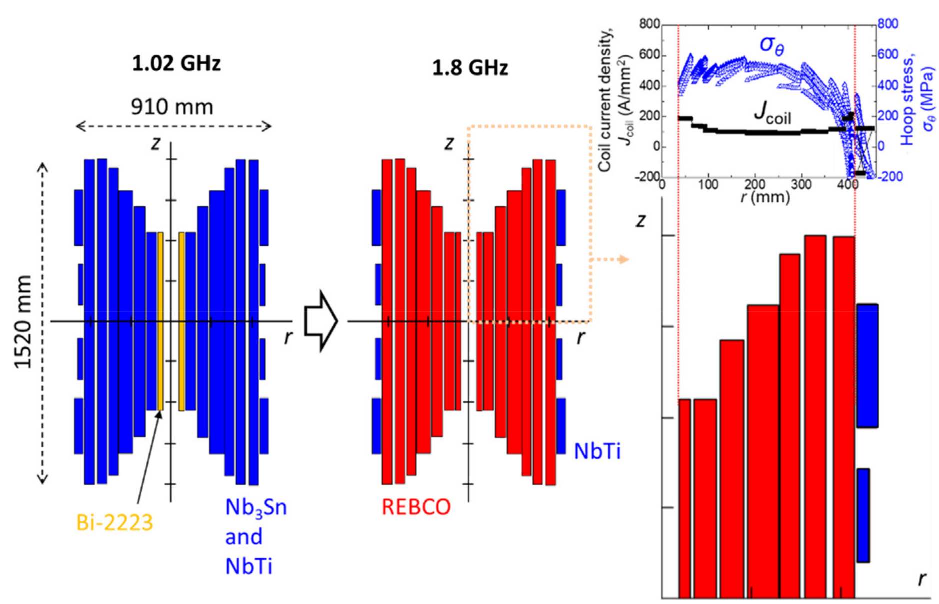

Fig. 5. An example of a preliminary design of the 1.8 GHz LTS/REBCO NMR magnet. The red-colored area corresponds to the REBCO coil, while the blue-colored area to the LTS coils. In the upper part, the hoop stress (σ_θ) due to the electromagnetic force is shown by the blue open triangles and the overall current density (J_{coil}) is indicated by the black line. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

cryogenic engineering enables a cryogen-free superconducting magnet using a cryocooler; in this case, a superconducting coil is conduction cooled by a cryocooler. In fact, a cryogen-free 25 T LTS/Bi-2223 magnet operated at 4–7 K has already been produced in 2016 [37]. However, such a cryogen-free cooling system is not applicable to the LTS NMR magnet, as the heat power necessary for persistent current switch (PCS) operation easily exceeds the cooling power of a cryocooler at 4 K. If the NMR magnet is made of HTS coils and operated in the persistent current mode, it can be operated at much higher temperature such as 10–20 K; as a 10–20 K cryocooler offers orders of magnitude higher cooling power than the 4 K cryocooler, a heat power for PCS operation becomes insignificant. As the upper critical magnetic field, H_{c2}, for an HTS wire decreases with the increase in temperature, the attainable magnetic field will be decreased to 700–800 MHz. From this point of view, a helium-free NMR magnet may be more suitable for lower field NMR magnets such as 200–500 MHz NMRs. Such HTS NMR magnets have already recently been produced using a driven mode HTS coil [38] or an HTS bulk [39].

3.5. Hybrid magnet

The NHMFL succeeded to develop a 35.2 T (1.5 GHz) NMR hybrid magnet [40] in 2017, which is comprised of a water cooled resistive inner magnet (22.2 T) and an LTS outer magnet (13 T); they are connected in series and charged simultaneously, thus resulting in low magnetic field fluctuation, 0.2 ppm, and high magnetic field homogeneity, 1 ppm. It is applicable to solid-state NMR, while not being sufficient for solution NMR. Although the hybrid magnet consumes electric power, it will be useful until the completion of a 1.5 GHz persistent mode LTS/HTS NMR magnet.

4. Summary

Based on an examination of the laboratory magnets and NMR magnets, the following future landscape is foreseen with regard to NMR magnets:

(a) The persistent mode 1.3 GHz LTS/HTS will be achieved by 2024. From the view point of HTS magnet technology, the development of a persistent mode 1.8 GHz LTS/REBCO NMR is possible, however both large amount of REBCO wires needed and its enormous cost are bottlenecks for realizing such a magnet.
(b) A helium-free persistent mode HTS NMR magnet will be available in the near future; it will be operated at 10–20 K, but its magnetic field limit may be only 700–800 MHz. A helium-free magnet is more suitable for lower field magnets such as 200–500 MHz.
(c) A hybrid magnet system for a 1.5 GHz NMR will be used until the completion of a persistent NMR magnet, although it has the drawback of large electric power consumption.

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