Some features of bulk melt-textured high-temperature superconductors subjected to alternating magnetic fields

P Vanderbemden¹, I Molenberg¹, P Simeonova² and V Lovchinov²

¹ University of Liège, Department of Electrical Engineering & Computer science (B28), Sart-Tilman, B-4000 Liège, Belgium
² Institute of Solid State Physics, Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee Blvd., 1784 Sofia, Bulgaria

E-mail: Philippe.Vanderbemden@ulg.ac.be

Abstract. Monolithic, large grain, (RE)Ba₂Cu₃O₇ high-temperature superconductors (where RE denotes a rare-earth ion) are known to be able to trap fields in excess of several teslas and represent thus an extremely promising competing technology for permanent magnet in several applications, e.g. in motors and generators. In any rotating machine, however, the superconducting permanent magnet is subjected to variable (transient, or alternating) parasitic magnetic fields. These magnetic fields interact with the superconductor, which yields a reduction of the remnant magnetization. In the present work we quantify these effects by analysing selected experimental data on bulk melt-textured superconductors subjected to AC fields. Our results indicate that the non-uniformity of superconducting properties in rather large samples might lead to unusual features and need to be taken into account to analyse the experimental data. We also investigate the evolution of the DC remnant magnetization of the bulk sample when it is subjected to a large number of AC magnetic field cycles, and investigate the experimental errors that result from a misorientation of the sample or a mispositioning of the Hall probe. The time-dependence of the remnant magnetization over 100000 cycles of the AC field is shown to display distinct regimes which all differ strongly from the usual decay due to magnetic relaxation.

1. Introduction

Due to their ability to carry high currents with no resistance at low temperature, superconducting materials represent a way of designing extremely promising electrical engineering applications [1-5]. In particular, (RE)Ba₂Cu₃O₇₋δ (REBCO) superconductors (where “RE” denotes a rare-earth ion) can carry extremely large currents under high applied fields, a property that can be exploited in long length coated conductors [6] as well as in current leads [7]. When prepared in the melt-processed form, e.g. monolithic disks of a few centimeters in diameter, the so-called “single domains” contain nanometer-sized defects that can act as efficient pinning centers for flux lines [8-11]. As a consequence, these materials are able to trapped considerable magnetic flux densities and act as powerful permanent magnets [12]. The world record of trapped field in such magnets [13] has been broken very recently [14] and is now 17.6 teslas, which is far beyond the saturation magnetization of conventional ferromagnetic materials. Such large magnetic flux densities give rise to huge Lorentz forces and considerable attention needs to be given to mechanical properties [13-15]. Another important aspect is related to the use of such permanent magnets in engineering applications. In running operation, the
remnant magnetization of bulk superconducting magnets is found to decrease slightly, which may be detrimental to the performances of the machine. The first origin of this decrease is magnetic relaxation, caused by a non-homogeneous magnetic flux profile (maximum in the center and zero at the sample edge) [16]. The corresponding time-dependence decay of magnetization, however, is usually logarithmic and for bulk REBCO samples at liquid nitrogen temperature (77 K), may become almost negligible after a few hours of operation [12]. The second cause of the decay is due to the interaction of the magnetized sample with transient or AC magnetic fields that may exist in the device. These time-dependent magnetic fields are due to vibrations of permanent magnets interacting with the superconductor (e.g. in magnetic bearings or magnetic levitation applications) or to variations of the rotating field generated by the stator windings (in rotating machines) [17,18]. Such parasitic alternating fields may reduce substantially the performance of the trapped field magnet [19]. In addition, the direction of the trapped field is also an important parameter, as described below.

Figure 1 shows four basic configurations of a bulk melt-textured superconductor subjected to an AC field. In this drawing, the bulk superconductor is assumed to be a cylindrical disk with the crystallographic $c$-axis perpendicular to the largest circular face ($ab$ planes). The AC magnetic field, can be applied to a virgin sample (configurations ‘a” and “b”) or to a sample in which permanent supercurrents, flowing in the $ab$ planes and generating a DC magnetization parallel to the $c$-axis (configurations “c” and “d”). The first two cases are traditional configurations in studying the AC magnetic response of a superconductor, e.g. by AC susceptibility. The AC magnetic field produces losses that, according to the Bean model in a homogeneous superconductor [16], are usually maximum when the AC currents induced by the field reach the sample center [20]. In large bulk samples, due to their relatively poor thermal conductivity [21,22], such losses may cause significant self-heating [23,24] which, in turns, has a detrimental effect on the critical current density $J_c$ of the superconductor. In the presence of a pre-existent DC magnetization (configuration “c”), such self-heating is present and yields some decrease of the trapped flux [19], which is dependent on the cooling conditions of the superconductor [24], e.g. through its surface/volume ratio [25] or the presence of a reinforcement stainless steel ring [26]. When the AC field is applied perpendicularly to the DC magnetization (configuration “d”), the decay of trapped flux is found to be severe: this corresponds to the so-called “crossed field” configuration which may lead to the “collapse of magnetic moment” [27]. This intriguing effect, studied for more than three decades [28], has been explained by considering the modification of current distribution caused by the varying field [29-31] but it still a subject of active research, see e.g. the recent work [32] on the crossed-field effect on stacks of tapes [33]. Similarly, the behaviour of a bulk melt-textured sample subjected to a large number of AC transverse cycles is difficult to predict but plays a significant role in engineering applications.

![Figure 1. Schematic diagram of a bulk melt-textured superconductor, showing the $ab$ planes and the $c$-axis. Four different configurations are illustrated: (a) AC field parallel to the $c$-axis, (b) AC field perpendicular to the $c$-axis, (c) sample with trapped DC magnetic moment (|| $c$) superimposed to a parallel AC field, (d) sample with trapped DC magnetic moment (|| $c$) superimposed to a transverse AC field ($\perp c$); the latter corresponds to the so-called “crossed-field” configuration.](image-url)
The purpose of the present work is to analyze several situations where a bulk melt-textured sample is subjected to an AC magnetic field and to point out some features that need to be taken into account for appropriate experimental characterization of these effects or interpretation of the measured data.

2. Experiment

The experiments are carried out on bulk DyBa$_2$Cu$_3$O$_7$ single domains synthesized by a melt-growth process described in detail in an earlier work [11]. Single grain samples up to 20 mm in diameter can be routinely fabricated by this technique. The critical superconducting transition temperature of the material is $T_c \sim 89$ K and its critical current density $J_c(77$ K, 0.1 T) $\sim 10^4$ A/cm$^2$. Several rectangular specimens intended to AC magnetic measurements were excised from the main single domain using a wire saw. Measurements of AC magnetic properties of cubic samples ($3 \times 3 \times 3$ mm) for $H \parallel c$ were carried out as a function of temperature in an AC susceptometer based on a cryocooler [34]. The crossed-field effects were investigated at liquid nitrogen temperature (77 K) on a parallelepipedic sample of dimensions $15 \times 15 \times 7$ mm, where the shortest dimension is parallel to the $c$-axis. The full penetration fields of this sample for $H \parallel c$ and $H \parallel ab$ were determined to be respectively $\mu_0 H_p \parallel c = 0.35$ T and $\mu_0 H_p \parallel ab = 0.25$ T. A field-cooled (FC) procedure under a constant 0.6 T background field produced by an iron-cored electromagnet was used to magnetize permanently the superconductor. The AC magnetic field was generated by a copper coil, fed either by a computer controlled power supply (quasi-static variations, frequency $f \sim 0.02$ Hz) or by a waveform synthesiser followed by an audio amplifier ($f \sim 25$ Hz). Measurements in the crossed-field configuration required to design a bespoke experimental set-up [29] adapted to large samples (~20 mm diameter, 5 mm thickness). Importantly, the sample was clamped firmly to prevent the strong magnetic torque (arising from the magnetic moment $\parallel c$ and the field $\parallel ab$) from modifying the sample orientation. The $c$-axis component of the DC average flux density was measured through numerical integration of the induced e.m.f. across a pick-up coil closely wound around the sample, while the local flux density was measured using an AREPOC Hall probe stuck against the sample surface.

3. Results

In a first set of experiments, the AC magnetic properties of selected samples was characterized in the absence of DC magnetization (configuration “a” in figure 1). Then crossed-field effects were investigated (configuration “d” in figure 1).

![Figure 2. Out-of-phase component of the AC magnetic susceptibility $\chi''$ vs. temperature measured on two samples of bulk melt-textured DyBa$_2$Cu$_3$O$_7$ superconducting sample for several amplitudes of the AC magnetic field. The applied AC field is parallel to the $c$-axis with no DC magnetization (configuration “a” in figure 1). The amplitude ranges from 5 gauss (0.5 mT) to 80 gauss (8 mT). The frequency is fixed at 1053 Hz.](image-url)
3.1. AC magnetic susceptibility in the absence of AC field

The magnetic response of melt-textured DyBa$_2$Cu$_3$O$_7$ samples was first studied as a function of temperature in order to examine the characteristics of the superconducting transition. Pick-up coils wound around the sample allow the AC magnetic susceptibility $\chi = \chi' - j\chi''$ to be determined [34,35].

Figure 3 shows the temperature dependence of the out-of-phase component $\chi''$ at several AC field amplitudes for two selected samples extracted from same the single domain. As can be seen, sample (a) displays a well-defined single peak at the smallest field investigated and a narrow superconducting transition at $T_c \sim 89$ K. The peak is shifted to lower temperatures and its amplitude increases as the AC field is increased, as expected from the Bean model taking into account a field-dependence of the critical current density [20]. In contrast, sample (b) displays a larger superconducting transition and a well-defined shoulder that can be viewed as the convolution of two separate peaks that occur close to each other. A double peak structure was often observed in high-temperature superconductors and attributed to shielding currents circulating either within the grains (intragranular) or across the grain boundaries (intergranular). Previous works on similar melt-textured samples have shown, however, that naturally occurring grain boundaries are characterized by a very small intergranular critical current density [36] (in contrast to artificially engineered boundaries [37]). The behaviour depicted in figure 3(b) is therefore likely to be exclusively of intragranular origin. A possible explanation would be the nucleation of secondary grains of smaller size [38] or local variations of oxygen content, resulting in an inhomogeneous distribution of $T_c$ in this particular sample. The latter hypothesis would be consistent to explain the unusual (and reproducible) feature occurring in the two peaks of the AC susceptibility: on increasing the AC field amplitude, the left peak is shifted to lower temperatures while the right main peak is shifted first to left (up to 60 G) and then to right (from 60 G to 100 G). Such a behaviour cannot be explained by the Bean model with a uniform $J_c$ and is expected to arise from a peculiar field-dependent $T_c$ and $J_c$ distribution within this particular sample.

This result underlines the possible occurrence of small $T_c$ and $J_c$ inhomogeneities when investigating the properties of medium-size (> 1 mm) melt-textured samples. The data displayed in figure 3(b) show that such inhomogeneities can be revealed easily through AC susceptibility measurements. Therefore considerable caution need to be taken before interpreting the results; the best specimens with sharp transition – cf. figure 3(a) – should be selected carefully before proceeding to further experiments.
Two examples of crossed-field experiments on a bulk melt-textured DyBa$_2$Cu$_3$O$_7$ superconducting sample illustrating the influence of experimental parameters on the measured data. (a) Comparison between the results obtained with a correctly positioned Hall probe (blue) or when the sample is tilted at 4° (red). (b) Comparison between the results obtained with a centred Hall probe (red) or when the Hall probe is placed near the edge of the sample (blue).

3.2. AC field applied perpendicularly to pre-existing magnetization

Now we investigate the results obtained in the “crossed field” configuration. A typical result of the decay of the $c$-axis average magnetic flux density against the transverse field AC field is plotted in figure 3. This graph shows how the average flux density (probed by the sensing coil) is affected drastically when cycles of transverse field are applied to the single domain. This behaviour is in excellent agreement with other results of crossed field experiments carried out on other superconducting materials [27-31]. It should be emphasized that the DC signal plotted in figure 3 is the average flux density, which may differ from the true magnetization due to finite-size effects, as shown clearly in ref. [39]. On figure 4, the results of similar experiments are displayed, but what is plotted is the central flux density against the surface of the sample, as recorded by a miniature Hall probe stuck against the sample surface. Two experiments were carried out in order to investigate the influence of parameters of the experimental system on the measured data. On figure 4(a), the Hall probe was intentionally misoriented by a small angle (4 degrees) while in figure 4(b) the Hall probe was purposefully placed near the edge of the sample. The results are always compared to those obtained with a correctly placed Hall probe. The two set of curves plotted in figure 4(b) are very close to each other, showing that the exact positioning of the Hall probe against the surface is not a critical parameter of the experiment. Figure 4(a), however, shows that a correct Hall probe angular orientation is essential. Following similar sets of experiments carried out at different angles, it was determined in practice that the (unavoidable) experimental misorientation that can be tolerated should be 0.5° or less.

It is of interest to investigate what happens to the sample DC signal when a large number of such transverse AC field cycles are applied, and to compare the resulting decay to that caused by magnetic relaxation effects only. The result of such experiment is plotted in figure 5, where the permanently magnetized superconductor was subjected to either 10 000 or 20 000 transverse AC field cycles. After application of 10 000 cycles (white symbols), the magnetic relaxation occurs but the corresponding decay is hardly perceptible. On the contrary, the decrease of $c$-axis flux density caused by further applications of transverse field cycles (black symbols) is found to be much more significant than the
Figure 5. Log-log plot of the decrease of the c-axis component of the DC central flux density (normalized to its initial value) of a bulk melt-textured DyBa$_2$Cu$_3$O$_7$ superconducting sample as a function the number $N$ of cycles of transverse AC field (parallel to the $ab$ planes). The measurement is carried out at $T = 77$ K. In the two experiments, either 10 000 cycles or 20 000 cycles are applied. The inset compares the magnetization decay in the two situations.

decrease caused by magnetic relaxation (flux creep). The reason for this behaviour is also due to the modification of the flux creep regime after the transverse field cycles are applied [40]. Further investigation of such effects using field amplitudes much smaller than the full-penetration field – as is the case in rotating machines – was carried out very recently [41].

Figure 6 shows the results of the application of more than transverse 100 000 cycles. Interestingly, two distinct power law regimes are observed, respectively for $N < 500$ and $N > 5000$. The remarkable feature of the decay observed in figure 6 is the failure of fitting the experimental data by any kind of exponential curve, as evidenced from the red line in figure 6. A possible hypothesis for the faster decay for the first 500 sweeps would be an initial decrease of the critical current density $J_c$ due to self-heating generated by the transverse field. Such magneto-thermal effects, studied numerically for superconducting cylinders of finite size [42] and analytically for infinite cylinders [24], are however unable to account for the behaviour observed in figure 6. Recent experiments [41] point out that subtraction of the extrapolated flux creep effects on the experimental data is helpful for the analysis, and demonstrate in fact that saturation of the magnetization due to AC transverse fields.

Figure 6. Log-log plot of the decrease of the c-axis component of the central DC flux density (normalized to its initial value) of a bulk melt-textured DyBa$_2$Cu$_3$O$_7$ superconducting sample as a function the number $N$ of cycles of transverse AC field (parallel to the $ab$ planes) for a large number of cycles. The measurement is carried out at $T = 77$ K. The blue and green lines are power law fits that are characteristic of the two regimes. The red line shows an attempt to fit the experimental data between 100 and 100 000 cycles by an exponential law.
can be observed when the number of cycles is raised by a further decade. Such a result is extremely important for use of bulk melt-textured superconductors in engineering applications.

5. Conclusion
The superconducting properties of bulk melt-textured DyBa$_2$Cu$_3$O$_7$ samples have been studied under various configurations of AC magnetic fields. The results point out that the non-uniformity of superconducting properties ($T_c$ and $J_c$ distribution), even in millimetre size samples, may lead to unusual features in the superconducting transition. We have shown that the application of an AC field to a pre-existing magnetization requires a carefully designed experimental set-up in which the orientation of the Hall probe with respect to the sample plays a crucial role. Finally, the decay of the DC flux density was observed for a large number of AC field cycles, and the corresponding data were shown to exhibit power law behaviour. Two distinct regimes have been pointed out, but they markedly differ from flux creep effects.

Acknowledgements
We thank the University of Liège (ULg) and the Ministry of Higher Education of Communauté Française de Belgique for a research grant Action de Recherches Concertées (ARC 11/16-03). We thank Profs. M. Ausloos, H. Caps, B. Vanderheyden, as well as Dr. J.F. Fagnard and P. Laurent for many fruitful discussions. The help of Dr. J. P. Mathieu and Prof. R. Cloots for supplying the superconducting sample is also acknowledged. This work is part of a collaboration programme financed by Wallonie Bruxelles International (WBI, Belgium) and the Bulgarian Academy of Sciences (BAS, Bulgaria).

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