Surface-Barrier Effects in the Microwave Second-Harmonic Response of Superconductors in the Mixed State

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Abstract. We report on transient effects in the microwave second-order response of different type of superconductors in the mixed state. The samples have contemporarily been exposed to a dc magnetic field, varying with a constant rate of 60 Oe/s, and a pulsed microwave magnetic field. The time evolution of the signal radiated at the second-harmonic frequency of the driving field has been measured for about 500 s from the instant in which the dc-field sweep has been stopped, with sampling time of \(\sim 0.3\) s. We show that the second-harmonic signal exhibits two relaxation regimes; an initial exponential decay, which endures roughly 10 s, and a logarithmic decay in the time scale of minutes. Evidence is given that the decay in the time scale of minutes is ruled by magnetic relaxation over the surface barrier.

PACS. 74.25.Ha Magnetic properties – 74.25.Nf Response to electromagnetic fields (nuclear magnetic resonance, surface impedance, etc.) – 74.60.Ge Flux pinning, flux creep, and flux-line lattice dynamics

1 Introduction

Measurements of magnetic relaxation in superconductors in the mixed state allow determining the rate of fluxons to overcome pinning and surface barriers \[\text{[4]}.\] Indeed, the interactions of fluxons with both the pinning and the surface barriers give rise to hysteretic behavior of the magnetization and, consequently, transient effects. The two barriers affect the magnetization curve in a different way \[\text{[4]}.\] In particular, surface-barrier effects manifest themselves in: i) first-penetration field, \(H_p\), higher than the lower critical field, \(H_{c1}\) \[\text{[4,13]}; ii) hysteresis loop of the magnetization curve asymmetric in the two branches at increasing and decreasing fields \[\text{[4,5,7,8]}; iii) magnetic relaxation rates different for flux entry and exit \[\text{[7,9,10,11,12]}\]. The effects of the surface barrier on the magnetization curve is conveniently investigated at temperatures near \(T_c\) where the bulk pinning is ineffective and, consequently, the asymmetry of the hysteresis loop due to the surface barrier can be highlighted. On the contrary, magnetic relaxation over the surface barrier is conveniently investigated at low temperatures, where the relaxation over the bulk-pinning potential is expected to occur at longer times.

In this paper, we investigate the second-order response of different superconducting samples, exposed to a sweeping dc magnetic field and a pulsed microwave (mw) magnetic field. It has been previously shown \[\text{[13,14]}\] that the mw second-order response of superconductors in the mixed state exhibits transient effects that, in the time scale of minutes, are characterized by variation rates different for dc magnetic fields reached at increasing and decreasing values. It has been hypothesized that these effects are due to motion of fluxons over the surface barrier \[\text{[13,14]}\]. The aim of the present work is to verify the validity of this hypothesis. To this purpose, we have investigated the time evolution of the mw response in superconducting samples characterized by the same bulk properties, but different quality of the surface through which the magnetic field penetrates. All the measurements have been performed at the liquid-He temperature, for different values of the dc field. After the sample has been exposed to a variation of the dc magnetic field, the signal radiated by the sample at the second-harmonic (SH) frequency of the driven field exhibits an initial exponential decay, which lasts about 10 s, and a logarithmic decay, in the time scale of minutes. The logarithmic-decay rate depends on the way in which the dc magnetic field has been reached, i.e. at increasing or decreasing values. Comparison of the results obtained in samples that differ only for the quality of the surface through which the magnetic field penetrates has shown that, in the time scale of minutes, the SH signal decays slower for smooth surface than for rough surface. These findings corroborate the hypothesis that, in this time scale, the time evolution of the SH signal is ruled by magnetic relaxation over the surface barrier.
2 Experimental and Samples

Time evolution of the SH signal has been studied in three samples of bulk ceramic MgB$_2$, and two samples of Nb polycrystal.

A sample of MgB$_2$ (which we indicate as Bo) has been extracted from a pellet sintered from Alfa-Aesar powder at 800°C in Ar atmosphere, for three hours. It has approximate dimensions $2 \times 1.5 \times 1$ mm$^3$ and $T_c \approx 38$ K. The largest faces of the sample correspond to the pristine surface of the pellet from which the sample was extracted; the other faces derive from the cutting of the pellet with a diamond saw. The faces have different roughness; those corresponding to the pristine surfaces of the pellet are smoother than the others.

Other two samples of MgB$_2$ (B1 and B2) have been extracted from a high-density (2.4 g/cm$^3$) pellet, which has been obtained by reactive infiltration of liquid Mg on a powdered B preform [15]. After the reaction in a sealed stainless steel container, lined with a Nb foil, a thermal treatment has been performed for two hours in the range of temperatures 850÷950°C. B1 and B2 samples have approximate dimensions $2 \times 3 \times 0.3$ mm$^3$ and $T_c \approx 39$ K. The largest faces of the B2 sample have been mechanically polished: they result much smoother than those of B1 sample.

The two samples of Nb (Nb1 and Nb2) have been extracted from the same batch, but the largest faces of Nb2 are much smoother than those of Nb1.

The sample is placed in a bimodal cavity, resonating at the two angular frequencies $\omega$ and $2\omega$, with $\omega/2\pi \approx 3$ GHz, in a region in which the mw magnetic fields $H(\omega)$ and $H(2\omega)$ are maximal and parallel to each other. The $\omega$-mode of the cavity is fed by a pulse oscillator, with pulse width 5 $\mu$s and pulse repetition rate 200 Hz, giving a maximal peak power of $\approx 50$ W (input peak power of the order of 10 W brings on microwave magnetic field of the order of 10 Oe in the region of the cavity in which the sample is located). A low-pass filter at the input of the cavity cuts any harmonic content of the oscillator by more than 60 dB. The harmonic signals radiated by the sample are filtered by a band-pass filter, with more than 60 dB rejection at the fundamental frequency, and are detected by a superheterodyne receiver. The cavity is placed between the poles of an electromagnet, which generates dc magnetic fields, $H_0$, up to $\approx 10$ kOe. All measurements here reported have been performed at $T = 4.2$ K with $H_0 \parallel H(\omega) \parallel H(2\omega)$.

Before any measurement was performed the sample was zero-field cooled down to $T = 4.2$ K; $H_0$ was increased up to 10 kOe and then decreased down to the residual field of the electromagnet. This preliminary procedure ensures that SH signals arising from processes occurring in weak links are suppressed by the trapped flux. The dc field was then swept with a constant rate of $\approx 60$ Oe/s up to fixed values and the evolution of the SH signal was measured for $\approx 500$ s from the instant in which each $H_0$ value has been reached, with sampling time $\approx 0.3$ s.

Fig. 1 shows the time evolution of the SH signal of sample Bo, measured from the instant in which $H_0$ has reached the value of 4 kOe, on increasing (circles) and decreasing (triangles) the field. Panel (a) shows the results obtained when both the dc and mw magnetic fields penetrate through the rough surfaces; panel (b) shows the results obtained when the fields penetrate through the smooth surfaces. During the first $\sim 10$ s the SH-vs-t curves of panels (a) and (b) show similar behavior. In this time scale, the SH signal shows an exponential decay. At longer times, the signal decays following a logarithmic law; moreover, the decay rate is different for the two orientations, being smaller when the fields penetrate through the smooth surfaces. Another peculiarity of the signal decay in the time scale of minutes concerns the decay rate of the SH signal after the sweep of the dc field has been stopped: the decay rate depends on the way the dc field has been reached (at increasing or decreasing values).

In order to deduce the parameters characteristic of the SH-signal decay, we have fitted the experimental data by the following expressions:

$$SH = A + B \exp \left(-t/\tau\right) \quad 0 < t < 10 \ s \quad (1)$$

$$SH = C[1 - D \log (t/t_0)] \quad t > 10 \ s \quad (2)$$

with $t_0 = 10$ s and $A = C$ the value that the intensity of the SH signal takes on at $t = t_0$.

The lines in Fig. 1 are the best-fit curves.
Measurements performed at different values of the dc magnetic field allowed us to determine the values of the best-fit parameters, $\tau$ and $D$, as a function of $H_0$. Fig. 2 shows the field dependence of the characteristic time of the exponential decay for the Bo sample. As one can see, $\tau$ is independent of the surface roughness and the way the dc magnetic field is reached; furthermore, it is roughly independent of $H_0$, within the experimental accuracy.

Fig. 3 shows the value of the best-fit parameter $D$ for the Bo sample as a function of $H_0$. The rate of the logarithmic decay depends on $H_0$ and its sweep direction; it is larger for magnetic fields reached at decreasing values than for fields reached at increasing values. On increasing $H_0$, the logarithmic-decay rates for negative and positive field variations approach each other. Furthermore, comparison between the results of panels (a) and (b) shows that the logarithmic-decay rate of the SH signal is about two times smaller when the magnetic fields penetrate through the smoother surface, in all the range of fields investigated.

Fig. 4 shows the time evolution of the SH signal for B1 (a) and B2 (b) samples, at $H_0 = 5$ kOe reached on increasing (circles) and decreasing (triangles) the field. It is worth to remember that B1 and B2 samples have the same bulk properties, but the faces of B2 sample have been polished and are much smoother than those of B1 sample. Comparison between the results of panels (a) and (b) shows that in the sample with polished faces the SH signal is roughly steady, in the time scale investigated. In the inset of panel (a) we report the field dependence of the logarithmic-decay rate of the SH signal, obtained by fitting the SH-vs-$t$ curves of B1 sample by Eq.(2). By fitting the initial decay of the SH signal of B1 sample by Eq.(1) we have obtained values of the best-fit parameter $\tau$ close to those obtained in the Bo sample (see Fig. 2).

In Fig. 5 we show the time evolution of the SH signal for Nb1 (a) and Nb2 (b) samples, at $H_0 = 4$ kOe, reached on increasing (circles) and decreasing (triangles) the field. It is worth to remember that the two samples differ for the quality of the surfaces, as indicated in the figure. Again, the SH signal of the sample with smooth surface decays slower than that of the sample with rough surface.

By fitting the SH-vs-$t$ curves of Nb samples, we have obtained values of the best-fit parameter $D$ one order of magnitude smaller than those obtained in the MgB$_2$ samples. Due to the slow variation, the logarithmic-decay rate of the SH signal in the Nb samples can be determined only with a large uncertainty. This finding does not allow comparing quantitatively the results obtained for $H_0$ reached on increasing and decreasing fields. Fig. 6 shows a comparison between the $D$ values obtained in the two Nb samples, at increasing (a) and decreasing (b) fields.

A further peculiarity of the SH response, which can be seen in the figures, is its hysteric behavior. As one can see, for the samples in which the SH signal shows a noticeable time evolution, the intensity of the signal is different for $H_0$ reached at increasing and decreasing fields. We infer that the hysteresis is strictly related to the transient effects.
$T_c$, the peculiarities of the SH signal are not significantly affected by the temperature; in particular, $\tau$ and $D$ take on values of the same order of magnitude as those obtained at $T = 4.2$ K. Only at temperatures very close to $T_c$ the SH signal is stationary and no hysteresis is observed.

3 Discussion

Nonlinear electromagnetic response of both conventional and high-$T_c$ superconductors has been discussed by different authors \cite{16, 17, 18, 19, 20, 21, 22, 23, 24, 25}. Several mechanisms give rise to emission of signals oscillating at harmonic frequencies of the driving field. At low magnetic fields and low temperatures, harmonic generation has been ascribed to nonlinear processes in weak links, impurities and intergrain-fluxon dynamics \cite{17, 18, 19, 20}. At magnetic fields higher than the lower critical field, when the weak links are decoupled, harmonic emission has been ascribed to intragrain-fluxon dynamics \cite{21, 22}. At temperatures near $T_c$, modulation of the order parameter by the em field is the main source of nonlinearity \cite{23, 24, 25}. To our knowledge, none of the models reported up to now in the literature discuss relaxation phenomena in the em response.

At low temperatures, after the samples have been exposed to magnetic fields larger than the first-penetration field, it is reasonable to hypothesize that a critical state develops. Nonlinear magnetization of superconductors in the critical state has been for the first time studied by Bean \cite{21}. The Bean model accounts quite well for the nonlinear response of conventional superconductors to low-frequency em fields. It is based on the hypothesis that the

Fig. 4. Time evolution of the SH signal for B1 (a) and B2 (b) samples, at $H_0 = 5$ kOe, reached on increasing (circles) and decreasing (triangles) the field. $T = 4.2$ K; input peak power $\approx 4$ W. Symbols are experimental data; lines are the best-fit curves obtained as explained in the text. Inset of panel (a) shows the field dependence of the best-fit parameter $D$ (lines are leads for eyes).

Fig. 5. Time evolution of the SH signal for Nb1 (a) and Nb2 (b) samples, at $H_0 = 4$ kOe, reached on increasing (circles) and decreasing (triangles) the field. $T = 4.2$ K; input peak power $\approx 4$ W. Symbols are experimental data; lines are the best-fit curves obtained as explained in the text.

Fig. 6. Logarithmic decay rate, $D$, of the SH signal of the two Nb samples, as a function of $H_0$ reached at increasing (a) and decreasing (b) fields. Lines are leads for eyes.
critical current does not depend on the magnetic field; furthermore, it tacitly assumes that the fluxon lattice follows adiabatically the em field variations. On these hypotheses, the response of the sample is even during the period of the em field; consequently, only odd-harmonic emission is expected. Even harmonics can be expected by taking into account the field dependence of the critical current according to the Anderson and Kim critical state model. However, for \( H_{dc} \gg H_{ac} \) the results obtained by using the Anderson-Kim model converge to those of Bean and only odd harmonics are expected. Nevertheless, it has been reported that superconductors in the critical state exposed to pulsed mw fields exhibit odd as well as even-harmonic emission, even when \( H_{dc} \gg H_{ac} \). The mw SH emission by superconductors in the critical state has been discussed by Ciccarello et al., who have elaborated a phenomenological model, based on the Bean model, in which the additional hypothesis is put forward that superconductors in the critical state operate a rectification process of the mw input field. The process arises because of the inertia of the fluxon lattice to follow high-frequency fields. It has been supposed that, due to the rigidity of the fluxon lattice, the induction field inside the sample does not follow adiabatically the mw field variations, except when the variations bring about motion of fluxons in the surface layers of the sample. From this model, it is expected that superconductors in the critical state radiate stationary SH signals, whose intensity is independent of the magnetic-field-sweep direction.

In the framework of the above-mentioned models, time evolution of harmonic signals could arise if the hypothesis is done that the critical state evolves toward a thermal-equilibrium state, by flux creep. However, several experimental evidences disagree with this hypothesis. Our results show that the time evolution of the SH signal exhibits two distinct regimes of relaxation: i) an exponential decay, with characteristic time independent of the surface roughness and the magnetic-field-sweep direction; ii) a logarithmic decay, with characteristic rate that strongly depends on the surface roughness and the field-sweep direction. The exponential decay is characterized by times of the order of seconds; so, it cannot be ascribed to magnetic relaxation by creep processes, especially so at low temperatures. On the other hand, relaxation through the pinning barrier does not justify the different decay rates observed after increasing and decreasing fields for times of the order of minutes. The dependence of the decay rate on the roughness of the sample surface, through which the magnetic field penetrates, strongly supports the idea that the decay of the SH signal in this time scale is due to surface-barrier effects.

In spite of the clear experimental evidence that the logarithmic decay of the SH signal is ruled by magnetic relaxation over the surface barrier, up to now there is not any model to quantitatively justify the experimental results. Indeed, it is worth noting that neither the Bean model nor the Ciccarello model take into account the presence of the surface barrier. Although in the literature are reported many studies on the surface barrier, none of them concern the effects of the barrier on the nonlinear em response. It would be of great importance to understand if surface-barrier effects induce relaxation of the signal or they could be themselves source of second-order response. As shown by Clem, the main effect of the surface barrier is the asymmetric response of fluxons for increasing and decreasing fields. We suggest that, due to this asymmetry, the presence of the surface barrier may be source of SH emission. In order to have this effect the amplitude of the oscillating field should be high enough to nucleate (or annihilate) fluxons near the sample surface during the em-field period. In this case, time evolution of the SH signal can be expected because after the magnetic-field sweep is stopped the amplitude of the surface barrier increases. The variation rate of the SH signal should be related to the rate of variation of the surface barrier that, as suggested by Burlachkov, is expected different for flux entry and exit.

Our results rule out the possibility that the initial exponential decay of the SH signal is ascribable to surface-barrier effects. Magnetic relaxation having characteristic times of the order of seconds has been reported by different authors. The authors of Ref. have discussed the exponential decay of the dc magnetization, detected during the first \( \sim 10 \) s of the relaxation process in BSCCO(2223)/Ag tapes, considering diffusive motion of fluxons induced by the variation of the dc magnetic field. Kalisky et al. have reported several studies on the time evolution of the magnetic-induction field in BSCCO crystals. They show that, during the magnetic-field sweep, two different vortex states coexist, characterized by different values of the persistent current density ("high" and "low"). The high-persistent-current state has been defined as a transient disordered vortex state (TDVS), because it decays with time when the external field is kept constant. For increasing fields, the TDVS state is located near the sample edges; for decreasing fields, it is located in the interior of the sample. When the magnetic field is kept constant, the "break" between the two states moves with time toward the sample edges or center, depending on the way the magnetic field has been reached (at increasing or decreasing values). Though the flux configuration is different at increasing and decreasing fields, the characteristic time with which the TDVS evolves toward an ordered state does not depend on the magnetic-field-sweep direction; furthermore, it results of the order of seconds.

We think that the exponential decay of the SH signal, revealed in the time scale of seconds, is ascribable to processes similar to those discussed in Refs. In particular, we suggest that it is related to the following process. During the field sweep, the fluxons arrange themselves in a configuration incompatible with the critical state; as soon as the field sweep is stopped, a diffusive motion of fluxons sets in; the process ends when the flux density in the bulk reaches the appropriate value for the critical state.
4 Conclusions

We have reported on transient effects in the mw second-order response of different types of superconductors in the mixed state. The measurements have been performed in a time window of $\sim 500$ s, after exposing the sample to a sweeping dc magnetic field ranging from 0 to 10 kOe. We have shown that during the first seconds the SH signal decays exponentially, while in the time scale of minutes it shows a logarithmic decay. The characteristic time of the initial decay does not depend on the field-sweep direction and the roughness of the sample surface through which the magnetic field penetrates. On the contrary, the variation rate of the logarithmic decay strongly depends on the surface roughness and the way in which the field is reached. These findings provide evidence that the two regimes of decay arise from different processes. In particular, we have suggested that the initial decay is related to diffusive motion of fluxon, which occurs during the time in which the critical state develops, while the logarithmic decay arises from magnetic relaxation over the surface barrier. Further investigation is necessary to understand whether surface-barrier effects are source of the second-order response, or fluxon motion over the surface barrier induces relaxation of SH signals arising from known nonlinear processes.

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