A LETTERS JOURNAL EXPLORING THE FRONTIERS OF PHYSICS

Transient and magnetic hysteresis in the microwave second-order response of BKBO crystals in the critical state

To cite this article: M. Li Vigni *et al* 2000 *EPL* **51** 571

EUROPHYSICS LETTERS 1 September 2000

Europhys. Lett., **51** (5), pp. 571–577 (2000)

Transient and magnetic hysteresis in the microwave second-order response of BKBO crystals in the critical state

M. LI VIGNI, A. AGLIOLO GALLITTO and M. GUCCIONE

INFM, Unità di Palermo and Dipartimento di Scienze Fisiche e Astronomiche
Via Archirafi 36, I-90123 Palermo, Italy

(received 24 March 2000; accepted in final form 30 June 2000)

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

Abstract. – The second-order microwave response of $Ba_{0.6}K_{0.4}BiO_3$ crystals in the critical state is investigated. Two different effects have been highlighted in second harmonic emission by superconductors exposed to intense pulsed microwave fields: a hysteretic behaviour and a time decay of the second harmonic signal intensity. Evidence is given that the two effects are strictly related.

It has been widely shown that superconductors in the critical state, under the action of intense em fields, exhibit a magnetisation containing Fourier components at the harmonic frequencies of the driving field [1–5]. In this letter, we discuss a novel effect observed in the microwave second-order response of $Ba_{0.6}K_{0.4}BiO_3$ (BKBO) crystals in the critical state. We have observed a magnetic hysteretic behaviour of the second harmonic (SH) signals. We show that the occurrence of the hysteresis is related to the presence of a transient in the SH response, which arises when the fluxon lattice is driven by em fields of high frequency.

Non-linear magnetisation of type-II superconductors in the critical state has been for the first time studied by Bean. The Bean model accounts quite well for the nonlinear response of conventional superconductors to low-frequency fields [1]. It is based on the hypothesis that the critical current does not depend on the magnetic field. Furthermore, it is tacitly assumed that the fluxon lattice follows adiabatically the em field variations. On these hypotheses, only odd harmonic emission is expected. Generation of even harmonics is expected by taking into account the field dependence of the critical current, according to the Anderson and Kim critical state model [6]. In this case, when dc and ac magnetic fields are simultaneously applied the fluxon lattice response to the positive and negative half-periods of the ac field will be different, with consequent even harmonic generation [2,3]. The latter model has been used to justify even harmonic generation in Josephson junctions in the critical state, submitted to ac and dc fields of the same order of magnitude. However, when $H_{\mathrm{dc}} \gg H_{\mathrm{ac}}$ the critical current is predominantly determined by H_{dc} and the fluxon response will be symmetric in the two half-periods of the cycle of the ac field. In this region, the results obtained by using the Anderson-Kim model converge to those of Bean and only odd harmonic emission is expected [2].

It has been reported that superconductors in the critical state exposed to intense pulsed microwave fields exhibit odd as well as even harmonic emission [4], even when $H_{\mathrm{dc}} \gg H_{\mathrm{ac}}$. Ciccarello *et al.* [4] have elaborated a phenomenological theory, based on the Bean model, in which the additional hypothesis that superconductors in the critical state operate a "rectification" process is done. The hypothesis arises from the fact that, because of the rigidity of the fluxon lattice, the induction field inside the sample does not follow adiabatically the variations of a high-frequency field, except when the variations involve motion of fluxons in the surface layers of the sample. It has been supposed that, for "direct critical state" developed by increasing fields, the induction flux does not vary during the positive half-period of the mw field, while it does during the negative half-period. The opposite occurs when decreasing fields develop a "reverse critical state". On this hypothesis, the response of the sample will be uneven during the wave period, with consequent odd as well as even harmonic emission. From this model, it is expected that superconductors in a critical state *à la* Bean radiate stationary SH signals independent of both the intensity and the sweep direction of the dc field. The model accounts quite well for the SH response to pulsed mw fields by $\mathrm{YBa_2Cu_3O_7}$ (YBCO) samples in the critical state [4]. We will show that, in order to account for the peculiarities of the SH signal observed in BKBO crystals, the model has to be generalised by properly considering the time response of the fluxon lattice to high-frequency fields.

We have performed measurements of harmonic emission in a crystal of BKBO in the critical state: the emission spectrum contains a SH component, whose intensity depends strongly on the magnetic field sweep direction. Measurements have been performed by exposing the sample to a static and an intense pulsed mw magnetic field. The crystal has a nearly cubic shape with edges of about 1 mm, it undergoes a superconducting transition at $T_{\mathrm{c}} \approx 30\,\mathrm{K}$. The sample is placed in a bimodal cavity, resonating at two angular frequencies ω and 2ω, with $\omega/2\pi \approx 3\,\mathrm{GHz}$, in a region in which the mw magnetic fields $\boldsymbol{H}(\omega)$ and $\boldsymbol{H}(2\omega)$ are maximal and parallel to each other. The ω-mode of the cavity is fed by a pulsed oscillator, with pulse repetition rate of 200 pps, pulse width $\approx 1.5\,\mu\mathrm{s}$ and maximal peak power $\approx 500\,\mathrm{W}$. The SH signals are detected by a superheterodyne receiver. All measurements have been performed with the static magnetic field $\boldsymbol{H}_0$ parallel to both $\boldsymbol{H}(\omega)$ and $\boldsymbol{H}(2\omega)$, at $T = 4.2\,\mathrm{K}$.

Figure 1 shows the SH signal intensity in the crystal of BKBO as a function of the static field. The curve has been obtained starting from $H_0 = 3\,\mathrm{kOe}$ and following the path indicated by the arrows. The intensity of the SH signal depends on the sweep direction of the magnetic field: at decreasing fields the signal is more intense than at increasing fields. For $H_0 > 1\,\mathrm{kOe}$ the amplitude of the hysteresis is independent of H_0 and of the spanned range; it is as well independent of the value of H_0 at which the inversion of the field sweep is operated. These peculiarities have been checked up to $H_0 \approx 10\,\mathrm{kOe}$.

We wish to remark that, during the first run to high fields, zero-field–cooled samples exhibit an intense low-field SH signal, which can be ascribed to non-linear processes occurring in weak links [7]. After exposition to high fields the sample goes into the critical state, the low-field signal disappears irreversibly and only a "remanent" signal is observed. Figure 1 shows just the behaviour of this remanent signal.

The hysteresis shown in fig. 1 is not accounted for by the model of ref. [4]. This is so because in both the reverse and direct critical state the assumption that the fluxon distribution does not at all change during one half-period of the em wave was put forward. The model can be improved by supposing that the fluxon lattice "attempts" to follow the mw field variations, by responding with characteristic times related to flux redistribution processes. In the direct

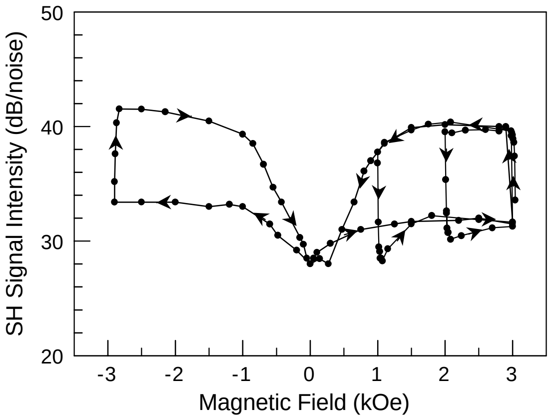

Fig. 1

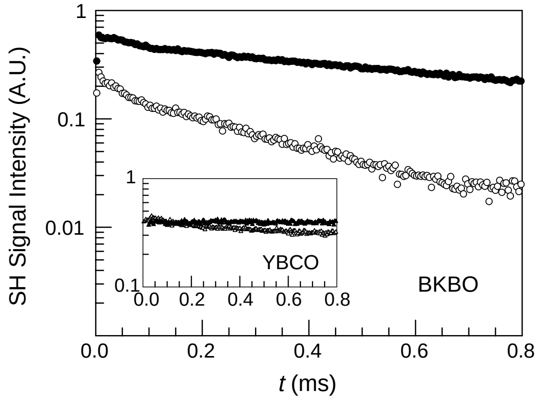

Fig. 2

Fig. 1 – SH signal intensity as a function of the static magnetic field in the BKBO crystal. The arrows indicate the followed path. $T = 4.2\,\mathrm{K}$; input peak power $\approx 80\,\mathrm{W}$.

Fig. 2 – SH signal intensity as a function of time, within the time interval of the mw pulse width, in the BKBO crystal. The inset shows the results in a YBCO crystal. $H_0 = 2\,\mathrm{kOe}$ attained increasing (open symbols) and decreasing (solid symbols) the external field. $T = 4.2\,\mathrm{K}$, input peak power $\approx 30\,\mathrm{W}$.

critical state one can suppose that during the positive half-period of the em wave the response time is much longer than the period of the wave, since the whole fluxon lattice has to move inward into the sample. During the negative half-period, in which only fluxons near the surface are involved, the response time is expected to be much shorter and the fluxons may follow adiabatically the mw field variations. The opposite is true in the reverse critical state. This leads to a partial rectification process, with the result that even harmonic emission is still expected. Furthermore, if the time response of the fluxon lattice is different for the direct and reverse critical state, the SH signals will exhibit hysteretic behaviour.

A partial rectification process gives rise to non-stationary SH signals during the time interval in which the mw pulse endures. This occurs because, under the action of the mw field, some fluxons will actually penetrate into the sample. At the beginning of two consecutive cycles, the induction field will be different. A more detailed explanation of the fluxon redistribution process during the mw pulse will be given later on in the paper. To check this expectation, we have exposed the sample to mw pulses lasting about 1 ms and we have measured the time dependence of the SH signal intensity, in the time interval in which the mw pulse endures. The mw pulses have been obtained by modulating a continuous wave of low intensity, frequency $\approx 3\,\mathrm{GHz}$, by a train of pulses $\approx 1\,\mathrm{ms}$ long with pulse repetition rate $\approx 1\,\mathrm{pps}$. The mw field, obtained in such a way, is amplified and used to feed the ω-mode of the bimodal cavity. The maximal peak power is $\approx 50\,\mathrm{W}$.

Figure 2 shows the SH signal intensity detected in the BKBO sample as a function of time. Measurements have been performed soon after H_0 had reached the value of 2 kOe, at increasing field (open circles) and at decreasing field (solid circles). The SH signal decays during the time in which the mw pulse endures. The decay times depend on the way the H_0 value has been reached: the signal decays faster when the H_0 value has been reached at increasing fields. For a comparison, we show in the inset the time dependence of the SH signal detected in a YBCO crystal. In this sample the SH signal does not decay in the reverse critical state (full triangles, above) and decays very slowly in the direct critical state (open triangles, below);

similar results have been obtained in a sample of ceramic YBCO. Measurements performed in the YBCO and BKBO samples at different values of H_0 have shown that: at $H_0 = 0\,\mathrm{SH}$ signals do not decay at all; for $H_0 < 0.5\,\mathrm{kOe}$ they decrease less fast than at higher fields; for $H_0 > 1\,\mathrm{kOe}$ the decay times are roughly independent of H_0.

As one can see from fig. 2, the time dependence of the SH signal intensity cannot be described by a single exponential law. Each curve shows an initial relatively fast decay followed by a slower one. The slow part may be described by $\exp[-t/\tau]$. By best-fitting this part, for the BKBO crystal we obtained $\tau = 0.32\,\mathrm{ms}$ at increasing H_0 and $\tau = 0.97\,\mathrm{ms}$ at decreasing H_0; while for YBCO $\tau = 4\,\mathrm{ms}$ at increasing H_0. The slope of the initial fast decay is at least 3 times larger. A transient response, with peculiarities similar to those we have observed, has been reported in mw power-absorption measurements in ceramic YBCO exposed to step-like changes of the static field [8]. The results concern measurements performed at low static fields, where processes involving Josephson fluxons come into play. The authors observed an initially rapid decay followed by a slower one. They ascribed the effect to transient shielding currents reflecting processes of magnetic flux redistribution. Our measurements have been performed at low temperatures and high magnetic fields; consequently, they may reflect relaxation processes of Abrikosov fluxons in the critical state. At high temperatures, where no critical state can be hypothesised, no decay of the SH signals is observed.

The transient response in the SH emission can be understood by considering what happens when a superconductor in the critical state is exposed to an abrupt field variation. Let us suppose that a superconducting sample in the direct critical state, developed by a dc field H_0, is exposed to a step-like change of the field of amplitude $\Delta H > 0$. Since the fluxons cannot instantaneously redistribute to a new critical state, a screening current will arise within the skin depth [8]. Such a current will decay and the fluxons will relax with a characteristic time τ toward a distribution appropriate to the direct critical state developed by a dc field $H_0 + \Delta H$. If, instead of the dc field step, an em field $H_1 \sin(\omega t)$ is applied, with $\omega \gg 1/\tau$, the fluxon lattice will relax toward a distribution suitable to a critical state developed by $H_0 + H_1 \sin(\omega t)$. What occurs during the mw pulse can be understood looking at the sequence shown in fig. 3.

Figure 3 (a) is a picture of the time evolution of the induction field profile soon after the mw pulse has been switched on (first cycle of the mw field and part of the second one). After the application of the pulse, some fluxons will penetrate into the sample with a rate related to the characteristic time τ. Such a process will continue as long as the field profile is that described by the dashed line, *i.e.*, when $B(0) = H_0 + H_1 \sin(\pi - \delta_1)$. Here, we neglect the decrease of the local field, from its surface value to its average interior value, within the fluxon-free region [9]; however, this does not affect the qualitative description of the process. The value of δ_1 decreases on increasing τ. For $\pi - \delta_1 \le \omega t \le 2\pi + \delta_1$ only the fluxons near the surface are involved in the motion; the field can follow the mw field variations. During the first part of the second cycle ($2\pi \le \omega t \le 2\pi + \delta_1$) the rectification process will not take place. For $\omega t > 2\pi + \delta_1$ again additional fluxons will penetrate up to $\omega t = 3\pi - (\delta_1 + \delta_2)$, with $\delta_2 \approx \delta_1$. In the subsequent cycles, on elapsing the time from the instant at which the mw pulse has been switched on, the rectification process, due to the uneven response of the fluxon lattice, will take place for a smaller and smaller part of the mw field period. This yields a decrease of the SH signal intensity.

We wish to remark that the AB line of fig. 3 (a) does not represent the effective time dependence of the field that the fluxon lattice follows. It indicates that at $t = (\pi - \delta_1)/\omega$ the field profile will be the one suitable to a direct critical state developed by a field $H_0 + H_1 \sin(\pi - \delta_1)$. Since the decay times of the SH signal are at least 10^4 times the mw field period, it is expected $\delta_1 \ll 2\pi$. Figure 3 (a) is out of scale for both the value of δ_1 and the slope of the AB line.

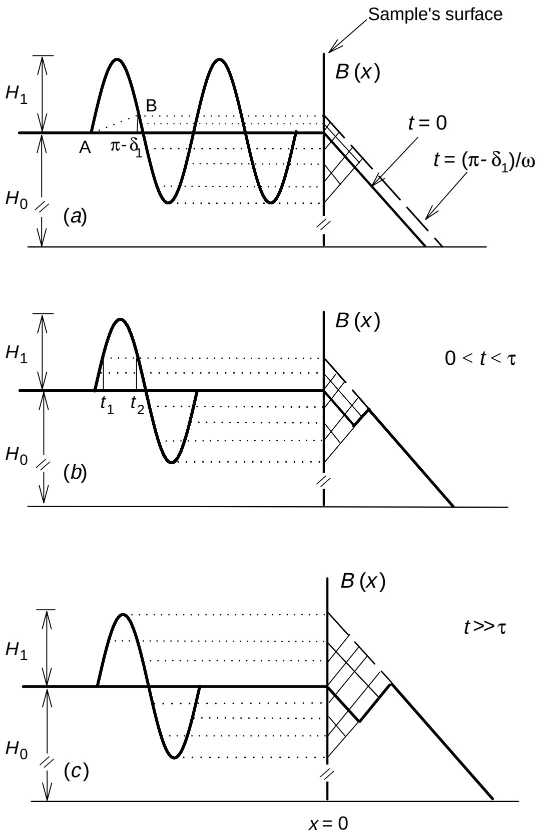

Fig. 3 – Time evolution of the induction field profile after the mw pulse has been switched on, as discussed in the text. (a) During the first cycle and part of the second cycle of the mw field; (b) during a generic cycle of the mw field occurring for $0 < t < \tau$; (c) after the steady state has been reached. The bold lines represent the field profile at the beginning of the cycles.

Figure 3 (b) shows the time evolution of the field profile during a generic cycle of the mw field which occurs for $0 < t < \tau$. The bold line represents the field profile at the beginning of the cycle. Now the fluxon lattice extends over a larger portion of the sample because additional fluxons have spread through. Up to $t = t_1$ the induction field can follow the mw field variations; for $t_1 < t < t_2$ the field profile will remain roughly unchanged; for $t > t_2$ it changes following again the mw field variations. Here t_1 and t_2 depend on τ and the time interval $t_2 - t_1$, in which the rectification process is effective, decreases on increasing the number of cycles.

Figure 3(c) shows what occurs at $t \gg \tau$. The magnetisation minor loop is symmetrical during the two half-periods of the wave. During the whole cycle, only the fluxons near the surface go in and out of the sample. No rectification process occurs and no SH signal is expected. So, when continuous-wave input fields are used no even harmonic generation is expected, independently of ω and τ, except for a transient initial signal.

The magnetic hysteresis of the SH signals arises whenever the response time in the direct critical state, $\tau_{\rm d}$, is different from the response time in the reverse critical state, $\tau_{\rm r}$. The results of fig. 2 show that the SH signal decay is steeper in the direct than in the reverse critical state.

This makes the rectification process more effective in the reverse critical state and justifies the larger SH signal intensity. It is expected that the larger is the ratio between τ_r and τ_d the larger is the amplitude of the hysteresis loop. Our results agree with the proposed model. In particular, at $H_0 = 0$ no decay of SH signal is observed and no hysteresis is detected; for $H_0 < 0.5\,\mathrm{kOe}$ only a slow decay is observed and the hysteresis has a small amplitude; for $H_0 > 1\,\mathrm{kOe}$ both the hysteresis amplitude and the decay times are roughly independent of H_0.

The validity of our idea is corroborated by the results obtained in the YBCO crystal (see the inset of fig. 2) where a very slow decay is observed. Accordingly, absolutely no, or very little, hysteretic behaviour has been observed in such a sample. Very likely, the hypothesis of complete rectification is legitimate since the response times of the fluxon lattice are extremely longer than the mw period. In a time scale of ms, both the hysteresis and the transient in the SH emission by the YBCO samples are not detected. In this case the model of ref. [4] is adequate in describing the experimental results.

The proposed model accounts, at least qualitatively, for the transient behaviour of the SH emission. The deviation from a mere exponential law of the decaying signals might be due to a distribution of relaxation times reflecting an inhomogeneous distribution of the pinning energies over the sample. However, we cannot *a priori* rule out the possibility that different relaxation processes come into play in different time scales.

Several authors have taken into account the surface barrier effects in discussing the hysteretic behaviour of the dc magnetisation and the ac magnetic permeability [10–12], as well as the relaxation of the dc magnetisation [13]. Burlachkov has shown that, because of the barrier effects, the flux entry in and exit from the sample can be very asymmetric [13]. He proved that the rate of flux entry is greater than that of flux exit. The author discusses the relaxation processes of the dc magnetisation from the critical state toward the thermodynamic equilibrium state. In this case, the relaxation occurs in a time scale much longer than that in which we detected the mw SH response. The flux redistribution, to which we ascribe the decay of the SH signal, is due to the relaxation of the screening current that arises by a fast variation of the applied field. Further investigation is necessary to make clear the surface barrier effects on the mw SH emission by superconductors in the critical state. At present, we have not a microscopic explanation for the different relaxation times of the fluxon lattice in the direct and reverse critical state. However, our results give evidence that the relaxation kinetics is slower when the fluxons are forced to leave than to penetrate the samples.

In conclusion, we have reported experimental results of SH emission in crystalline BKBO in the critical state exposed to intense pulsed mw fields. We have measured the time dependence of the SH signal intensity, in the time interval in which the mw pulse endures, for mw pulses $\approx 1\,\mathrm{ms}$ long. We have observed that the SH signal exhibits a time decay, which does not follow a single exponential law. The transient has been related to processes of magnetic flux redistribution, which come into play when the fluxon lattice is driven by high-frequency fields. Our results have shown that the time spent by the fluxons to rearrange in a suitable lattice for a critical state developed by the total field, dc plus ac, is longer in the reverse than in the direct critical state. Furthermore, a magnetic hysteretic behaviour of the SH signal has been found; the signal is more intense at decreasing than at increasing fields. Since the decay times of the SH signal are different in the direct and reverse critical state, the amplitude of the hysteresis loop depends on the time. The peculiarities of the hysteresis have been studied by exposing the sample to mw pulses $1.5\,\mu\mathrm{s}$ long. We have shown that the two effects, the hysteretic behaviour and the transient response, are strictly related to each other.

We have proposed a generalisation of the model of Ciccarello *et al.* in which the time response of the fluxon lattice is taken into due account. When the response times are much longer than the period of the driving field our model reduces to that of Ciccarello *et al.* On the contrary, if the response times are of the same order as the wave period, the fluxon lattice follows the em field variation and the results obtained from our model converge to those of the Bean model. Therefore, both the presence of even harmonic emission and/or the features of the SH spectra depend on the frequency of the driving field.

* * *

The authors acknowledge the staff of the ISSP of the Russian Academy of Sciences (Chernogolovka) for having supplied the BKBO crystal, and M. R. Trunin and I. Ciccarello for their continuous interest and helpful suggestions.

REFERENCES

[1] Bean C. P., *Rev. Mod. Phys.*, **36** (1964) 31.
[2] Ji L., Sohn R. H., Spalding G. C., Lobb C. J. and Tinkham M., *Phys. Rev. B*, **40** (1989) 10936.
[3] Muller K. H., Macfarlane J. C. and Driver R., *Physica C*, **158** (1989) 366.
[4] Ciccarello I., Fazio C., Guccione M. and Li Vigni M., *Physica C*, **159** (1989) 769.
[5] McDonald J., Clem J. R. and Oates D. E., *Phys. Rev. B*, **55** (1997) 11823.
[6] Kim Y. B., Hempstead C. F. and Strnad A. R., *Phys. Rev.*, **129** (1963) 528.
[7] Agliolo Gallitto A., Guccione M. and Li Vigni M., *Physica C*, **309** (1998) 8.
[8] Atsarkin V. A., Vasneva G. A., Demidov V. V. and Noginova N. E., *Solid State Commun.*, **74** (1990) 603.
[9] Clem J. R., in *Proceedings of the 13th Conference on Low Temperature Physics* (LT 13), edited by K. D. Timmerhaus, W. J. O'Sullivan and E. F. Hammel, Vol. **3** (Plenum, New York) 1974, pp. 102-106.
[10] Clem J. R., *J. Appl. Phys.*, **50** (1979) 3518.
[11] Chen D. X., Cross R. W. and Sanchez A., *Cryogenics*, **33** (1993) 695.
[12] Kugel K. I., Mamsurova L. G., Pigalskiy K. S. and Rakhmanov A. L., *Physica C*, **300** (1998) 270.
[13] Burlachkov L., *Phys. Rev. B*, **47** (1993) 8056.