

1-1-2008

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### Recommended Citation

METSKHVARISHVILI, IOSEB R. (2008) "Critical State Models for Intergrain Junctions of Polycrystalline Superconductors by Third Harmonic ac Susceptibility Measurements," *Turkish Journal of Physics*: Vol. 32: No. 2, Article 5. Available at: <https://journals.tubitak.gov.tr/physics/vol32/iss2/5>

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# Critical State Models for Intergrain Junctions of Polycrystalline Superconductors by Third Harmonic ac Susceptibility Measurements

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Received 11.09.2007

## Abstract

High harmonic response of high- $T_c$  and Chevrel-phase polycrystalline superconductors are measured in the presence of small ac excitation field and dc magnetic field applied on it. The aim of this work is to study the Josephson weak link behavior and compare the two systems in order to understand the granular nature of polycrystalline superconductors. Critical state models are used to explain the nonlinear magnetic response in polycrystalline superconductors.

**Key Words:** Low field, Josephson junction, Critical state models.

## 1. Introduction

Polycrystalline superconductors have a peculiarity associated with the existence of superconductivity in each grain, whose parameters are similar to a bulk single crystal. Between the grains there are weak Josephson junctions, which determine the superconducting properties of the whole system [1-4]. For the Josephson junctions in low fields, less than first critical field of the grains, a lot of irreversible and nonlinear phenomena are observed. These phenomena are characterized by the presence of high harmonics.

The aim of this work is to study the Josephson weak link behavior in high- $T_c$  and Chevrel-phase polycrystalline superconductors in weak magnetic fields and compare these two systems to understand granular nature of polycrystalline superconductors. Chevrel-phase superconductors are similar to the high-temperature oxide superconductors. In Chevrel-phase superconductors Ginsburg-Landau parameter  $\kappa$  is large ( $\sim 100$ ) and the coherence length  $\xi$  may be extremely short. Correspondingly, huge upper critical fields  $H_{c2} = 60$  T in  $\text{SnMo}_6\text{S}_8$  [4-5] was observed. It was a record value before the discovery of high-temperature oxide superconductors.

The high harmonics were first interpreted using Bean's critical state model [6], in which the amplitude of each harmonic is inversely proportional to the critical current density. It is a very good method for studying a distribution of the magnetic fields and currents in the critical state. The concept of the critical state introduced by Charles Bean is widely used to describe various physical phenomena in the vortex phase of type-II superconductors. In the original Bean model, the critical current density  $j_c$  was assumed to be independent from the local magnetic field and only odd harmonics were predicted. Unlike Bean's model, in

Anderson-Kim model it was found that even harmonics could be observed in the framework of the critical state model by taking into account the dependence of the field of critical currents [7].

## 2. Experiment

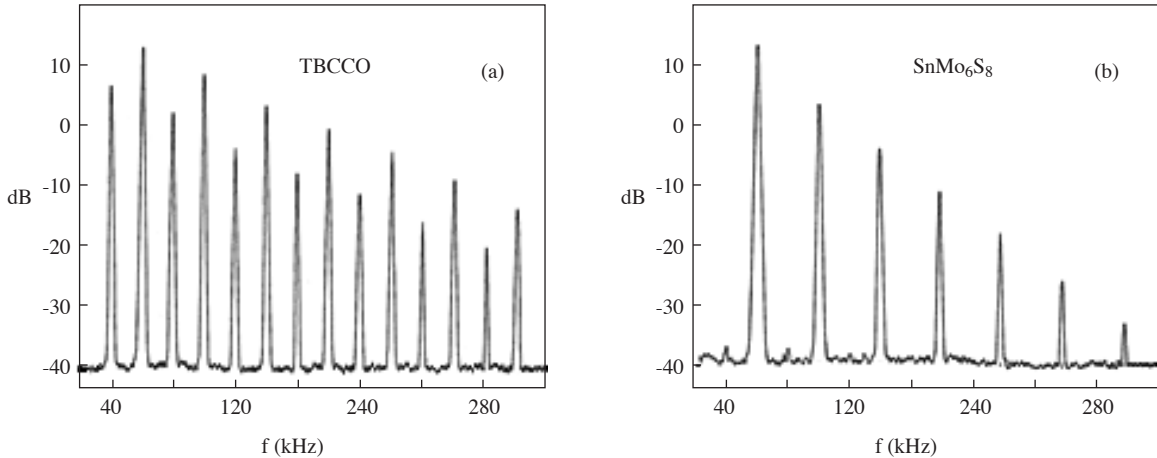
The investigated samples of  $Tl_2Ba_2Ca_2Cu_3O_y$  and  $SnMo_6S_8$  fashioned in pellet form of diameter  $\approx 9.5$  mm and thickness  $\approx 3.5$  mm. The samples were placed in a coil creating ac field  $h$  and the same coil was also used for measurements. Constant dc field  $H$  was parallel to ac field  $H(t) = H + h\cos\omega t$  and was created by external solenoid. Measurements have been performed mainly at 20 kHz frequency and in the range of fields  $10^{-2} \leq h \leq 1$  Oe,  $H \leq 12$  Oe.

The measuring system could be described as follows [8]. A sinusoidal current from the generator, which is injected in the induction coil containing the sample, excites an ac field. The response from the coil through the filter, which dampens the basic frequency, is transformed to a selective amplifier. The amplifier could be regulated by the desired number of harmonics. The signal from the selective amplifier is recorded with a voltmeter, which measures the amplitude of the harmonics. Since the odd harmonics are observed in both zero field and nonzero fields, even harmonics are observed only in case of an existing dc field. High harmonics spectra are observed via a spectrum analyzer.

Errors in high harmonics measurement might be as high as  $\sim 2\%$ , when the measuring signal is lower than  $0.2 \mu V$ . However, the error does not exceed 0.5% for higher amplitudes.

## 3. Results and Discussion

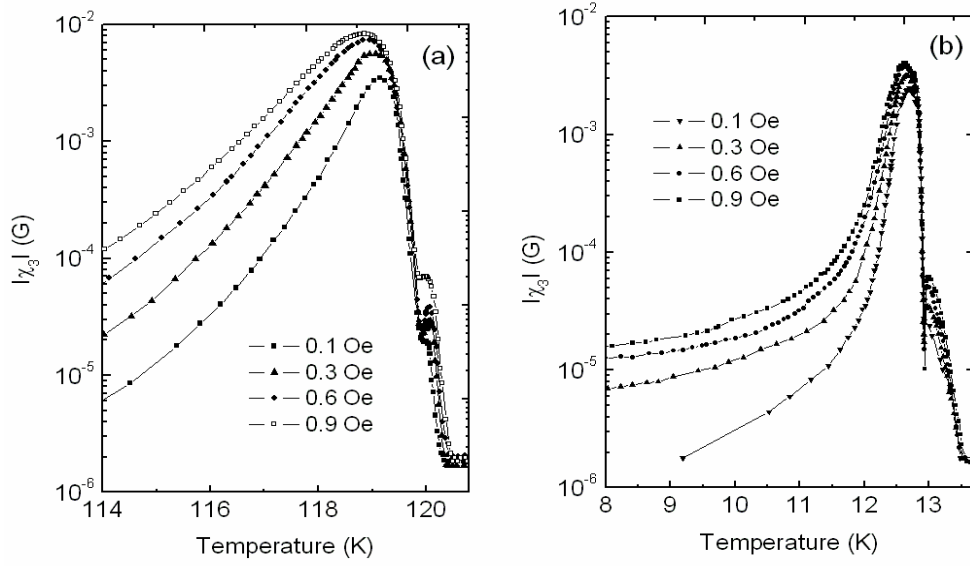
Figure 1 illustrates the spectrum of high harmonics measured for  $Tl_2Ba_2Ca_2Cu_3O_y$  and  $SnMo_6S_8$ , which was obtained in weak ac and dc magnetic fields. From Figure 1a it could be seen that the amplitudes of odd and even harmonics weakly decrease when the number of harmonic increases. Therefore, more reasonable is to use a low number of harmonics. For  $H = 0$  the shape of the odd harmonics does not change, while the even harmonics are actually vanished. By contrast, for  $SnMo_6S_8$  (see Figure 1b) even harmonics are absent for  $H \neq 0$ . Such experiments have been made in different range of the temperature. However, the experimental results are same and the even harmonics are not observed.



**Figure 1.** High harmonics spectrum: (a)  $Tl_2Ba_2Ca_2Cu_3O_y$ ,  $T \approx 118$  K,  $h = 0.6$  Oe,  $H = 8$  Oe; (b)  $SnMo_6S_8$ ,  $T \approx 12$  K,  $h = 0.6$  Oe,  $H = 8$  Oe.

The measurements for temperature dependence of the third harmonics for various ac magnetic field amplitudes at  $f=20$  kHz,  $H=0$  have shown that two-peaked curves are observed for HTSCs and SMS

(Figure 2). The effect can be explained by flux shielding between grains. The transition to superconducting states occurs at  $\approx 120.3$  K for TBCCO, and starts at  $T^* \approx 13.5$  K for SMS, which is typical for compounds in this class [4, 5]. When sample temperature is below  $T_c$ , the applied magnetic field is shielded from the superconducting grains and a low peak is observed. At lower temperature, the intergranular junctions start shielding the applied magnetic field and a second peak appears. Intragranular and intergranular peaks are very sensitive to the amplitudes of the applied field. As the ac magnetic field increased, the peaks shifted toward lower temperatures, becoming broader and increased monotonically. Via temperature dependence of the third harmonics it can be seen that, for SMS, this dependence turned out to be completely similar to that for ceramic TBCCO.



**Figure 2.** Temperature dependences of the amplitude of the third harmonics at 0.1, 0.3, 0.6, 0.9 Oe ac field and  $H=0$ , for (a)  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ , (b)  $\text{SnMo}_6\text{S}_8$ .

According to Bean's model [6], amplitudes of each harmonics are inversely proportional to the critical current density  $c_n \approx 1/j_c$ . Thus function  $j_c(B)$  can be determined by examining the dependencies of these harmonics on the magnitude of the constant field  $H$  at fixed amplitude of  $h$ .

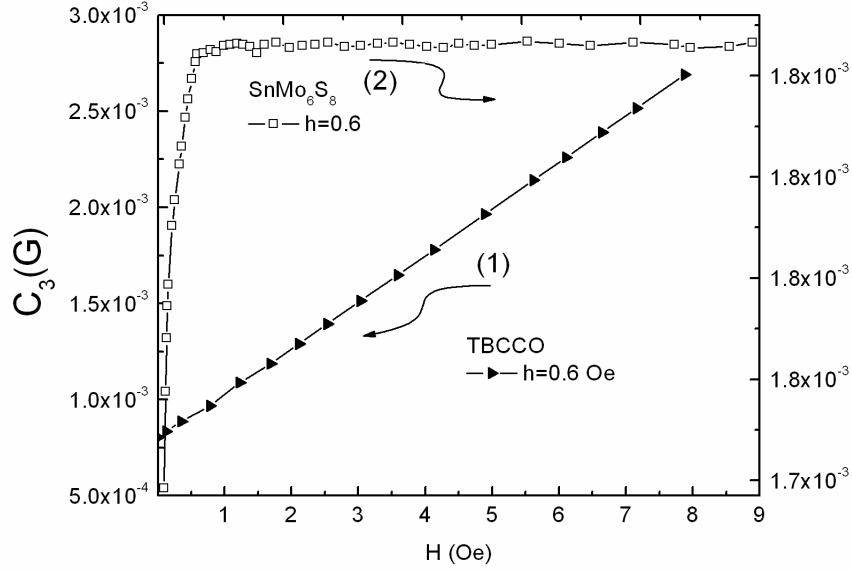
From Figure 3, it follows that, for TBCCO,  $c_3$  is very sensitive to the low external dc fields and increases linearly. The observed behavior of the third harmonic could be explained by the modal dependences between the values of critical current density and magnetic induction [7]:

$$J_c(B) = j_0 / (1 + B/B_0) \quad (3)$$

The same figure illustrates that the amplitude of the third harmonic for SMS does not depend on the magnetic field:

$$j_c(B) = j_0. \quad (4)$$

It must be noted that we carried out such experiments over a large temperature range and  $c_3(H)$  did not change for both samples.



**Figure 3.** Dependence of the third harmonic  $c_3$  on the dc-field. Curve 1 corresponds to  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ ,  $T \approx 118$  K,  $h = 0.6$  Oe, and curve 2 -  $\text{SnMo}_6\text{S}_8$ ,  $T \approx 12$  K,  $h = 0.6$  Oe.

## 4. Conclusion

Investigation under the same conditions show that nonlinear magnetic response from Josephson medium are quite different in polycrystalline samples of high- $T_c$  and Chevrel-phase materials. The high- $T_c$  materials are better described by Kim-Anderson model, where the critical current depends on the external magnetic field, and even harmonics are observed in dc fields. On the other hand, the Chevrel-phase materials follow the classical Bean critical state model, when the current does not depend on the field, and even harmonics are not observed.

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