Supercond. Sci. Technol. **9** (1996) 447–452. Printed in the UK

Radio-frequency magnetoabsorption in YBCO–Ag and BSCCO superconductors: the cross-over between two-level and one-level critical state models

Pratap Raychaudhuri†

Department of Physics and Meteorology, Indian Institute of Technology, Kharagpur, India

Received 17 November 1995, in final form 12 February 1996

Abstract. We have studied the virgin curve and hysteresis of radio-frequency magnetoabsorption in YBCO–Ag ($YBa_2Cu_3O_{7-\delta}$, $Ag_{1.2}$) and BSCCO ($Bi_2Sr_2CaCu_2O_{8+\delta}$) samples at 77 K in a magnetic field up to a value of 2 kOe. The hysteresis of YBCO–Ag is in good agreement with the prediction of the two-level critical state model. However, in the case of BSCCO the agreement is there only when the maximum field does not exceed 67.5 Oe for a frequency of 1.5 MHz. For higher fields the results resemble those of the one-level critical state model. This is due to the fact that at fields greater than 67.5 Oe the superconducting grain boundaries become normal. The cross-over point shifts to 55 Oe when the frequency is increased to 12 MHz. A qualitative explanation of this shift is given on the basis of the disordered model.

1. Introduction

Study of high-frequency loss is a convenient means of investigating the nature of the superconducting phase. It has been observed [1–3] that in the presence of a magnetic field the dependence on field of the surface resistance of high-temperature superconductors (HTSCs) is different from that in conventional type-II superconductors. The surface resistance of a zero-field cooled (ZFC) HTSC sample is lower in decreasing field than in increasing field and attains a minimum before the field comes to zero. In the case of conventional superconductors the surface resistance in decreasing field is higher than in increasing field and comes to a minimum only at negative values of field.

The hysteresis of surface resistance observed in homogeneous type-II superconductors has been explained in terms of the one-level critical state model. In this model the superconductor is characterized by a single critical current. In the mixed state the motion of fluxons is governed by the viscous drag and restoring force from the pinning centre. It has been shown that at high frequencies the viscous drag dominates [4], and the fluxons oscillate about their mean positions as if they were unpinned. Coffey and Clem [5] have shown that when viscous drag dominates, surface resistance R_s is given by

$$R_s = (\omega\mu_0/2)\sqrt{(2B_0\Phi_0/\mu_0\eta\omega)}$$

where ω is the frequency of the ratio-frequency (r.f.) field, B_0, the static magnetic induction inside the superconductor, η, the viscous drag as defined in the Bardeen-Stephen model [6], μ_0, the permittivity of free space and Φ_0, the flux carried by a fluxon. B_0 can be written as $B_0 = n\Phi_0$ where n is the fluxon density. Thus $R_s \propto \sqrt{n}$. Using the critical state model [7, 8] B_0 and hence n can be calculated. The typical nature of a curve calculated in this way is shown in figure 1(a).

Ji *et al* [1] used a two-level critical state model to explain the hysteretic behaviour of surface resistance in granular HTSC materials. This model takes into account the effect of granularity of these materials by assuming that the intragrain and intergrain regions are charecterized by different values of current. It assumes that on a macroscopic level the flux density averaged over a scale of many grains should have a gradient determined by the macroscopically flowing intergranular critical current density $J_{cj}(\sim 100\ \mathrm{A\ cm^{-2}})$ [9]. On the local level within single grains a critical state is established with a flux density gradient determined by intragranular critical current density $J_{cg}(\sim 10^4\text{–}10^5\ \mathrm{A\ cm^{-2}})$ [9], which is much larger than J_{cj}. The flux density in grain boundaries influences

---

† Current Address: Tata Institute of Fundamental Research, Homi Bhaba Road, Bombay 400005, India.

0953-2048/96/060447+06$19.50

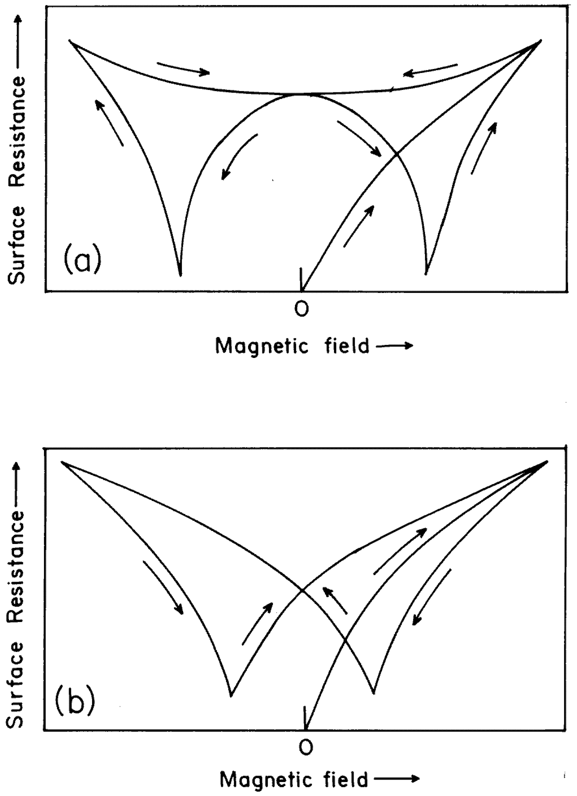

Figure 1. A schematic surface resistance hysteresis curve according to (a) the one-level critical state model and (b) the two-level critical state model (after Ji *et al* [1]).

this local critical state within the grains by supplying the effective external field. The effective flux profile is schematically shown in figure 2. It is further assumed that there are two types of fluxon, namely (i) the grain boundary fluxons, which pass only through the weakly superconducting intergranular regions and are relatively free (since the intergranular regions do not contain strong pinning centres), and (ii) the grain-pinned fluxons which are strongly pinned at the pinning centres inside the grains. In an r.f. it is the intergranular 'free' fluxons that contribute primarily to the loss. Thus $R_s \propto \sqrt{n_i}$, where n_i is the intergarnular fluxon density.

The surface resistance curve calculated using this model reproduces all the essential features of the observed surface resistance curve in HTSC materials, that is, the surface resistance in decreasing field is lower than in increasing field (as well as lower than the virgin curve; i.e. the surface resistance curve when the field is first swept from zero to the maximum value in the ZFC sample) and comes to a minimum before the field comes to zero (figure 1(b)).

In order to verify this model we performed measurements on silver-added YBCO samples ($YBa_2Cu_3O_{7-\delta}$, $Ag_{1.2}$) at frequency of 1.5 MHz and BSCCO ($Bi_2Sr_2CaCu_2O_{8+\delta}$) samples at frequencies of 1.5 MHz and 12 MHz. The results for YBCO–Ag samples followed the two-level critical state model up to the maximum applied field of 2 kOe (figure 4). However in the case of BSCCO there is a cross-over at 67.5 Oe between the increasing and decreasing field values of surface resistance, when the measurement is made at 1.5 MHz (figure 7). At 12 MHz this cross-over shifts to a lower value of field (figure 8). These data are explained on the basis of the critical state models.

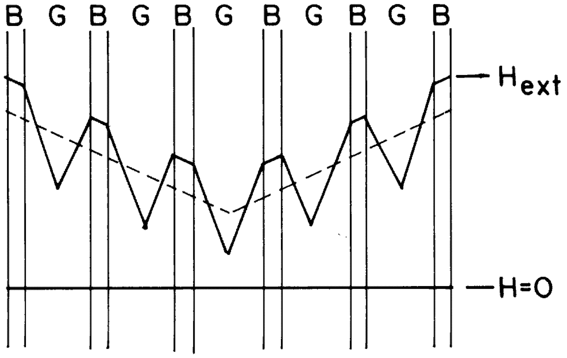

Figure 2. The two-level critical state: a schematic microscopic field profile (solid line) inside rains (Oe) and grain boundaries (B) and a macroscopic field profile (dashed line) inside the superconductor (after Ji *et al* [1]).

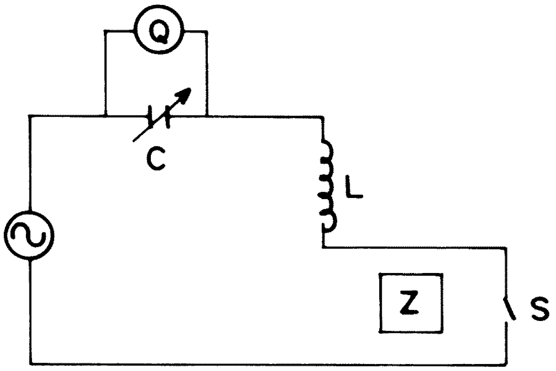

Figure 3. A block diagram of the *Q*-meter: C is the tuning capacitor, L is a standard inductor, S the shorting strip, and Z the coil inside which the sample is placed.

2. Superconducting samples and experimental procedure

$YBa_2Cu_3O_{7-\delta}$, $Ag_{1.2}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$ polycrystalline samples were prepared using the conventional solid state reaction route. The YBCO–Ag ceramic composite was prepared by calcining a mixture containing appropriate amounts of yttrium oxide and nitrates of barium, copper and silver at 920 °C in air for 40 h. The calcined mass was then ground, pelletized and sintered at 950 °C under flowing oxygen for 24 h, cooled to 500 °C and sintered for another 24 h under flowing oxygen. The sample was cooled at 1 °C min^{-1} under oxygen to room temperature. It has been shown by many groups [10, 11] that in YBCO addition of silver increases the itergranular coupling and stabilizes the oxygen content in the grain, thereby increasing its stability against environmental degradation in a humid

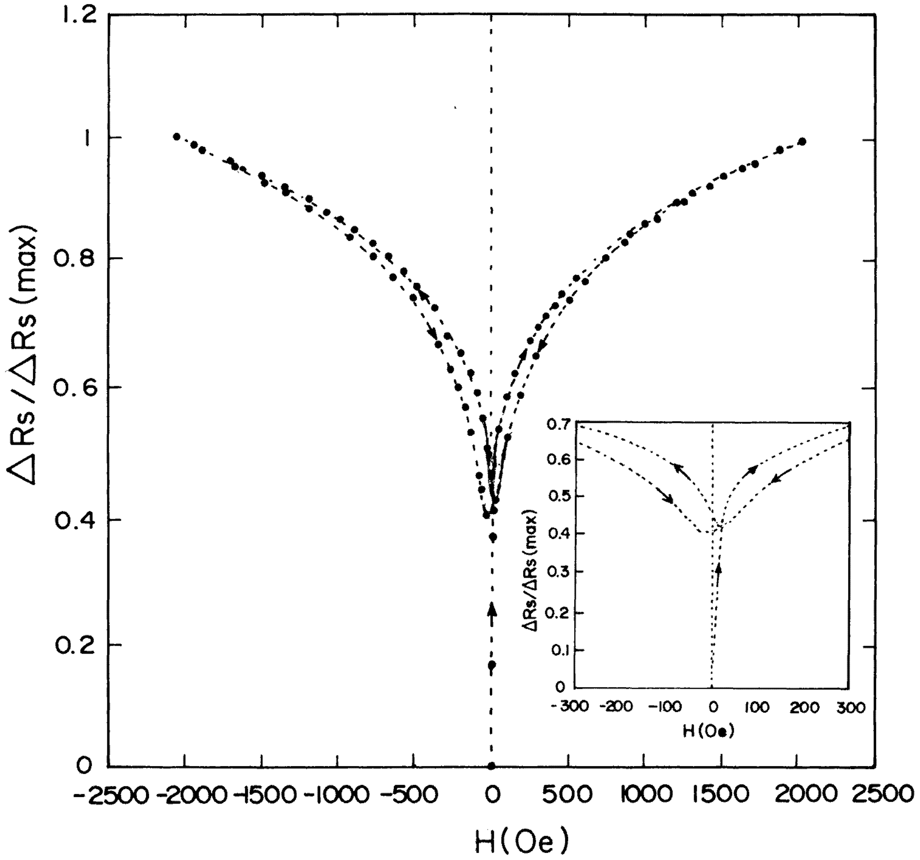

Figure 4. The surface resistance hysteresis curve of the YBCO–Ag sample up to a maximum field of 2000 Oe (at a frequency of 1.5 MHz); inset, an enlarged view.

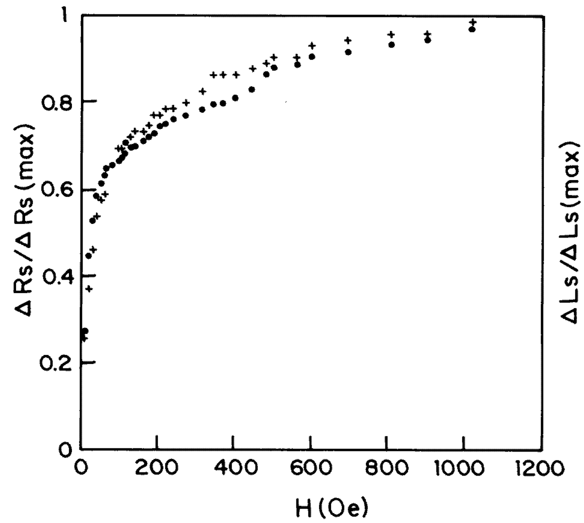

Figure 5. The virgin curve of surface resistance (●) and inductance (+) for the BSCCO sample (at frequency 1.5 MHz).

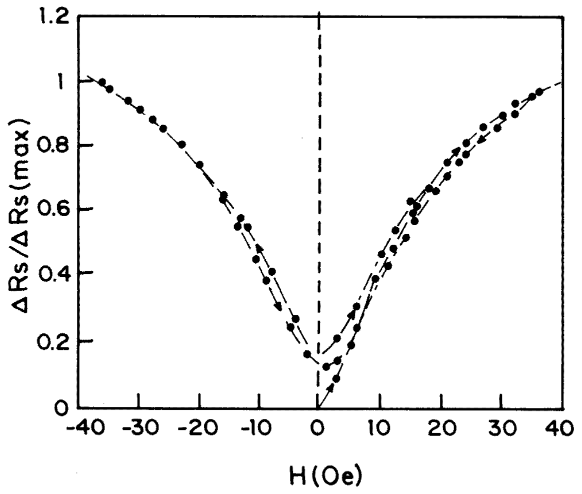

Figure 6. The surface resistance hysteresis curve of the BSCCO sample up to a maximum field of 40 Oe (at frequency 1.5 MHz).

atmosphere [12]. The BSCCO sample was prepared by mixing appropriate amounts of oxides of bismuth and copper and carbonates of calcium and strontium. This mixture was calcined at 850 °C for 24 h. It was ground, pelletized and sintered in air for 30 h at 800–900 °C and subsequently cooled to 500 °C and kept for 12 h. The sample was then quenched by dipping into liquid nitrogen.

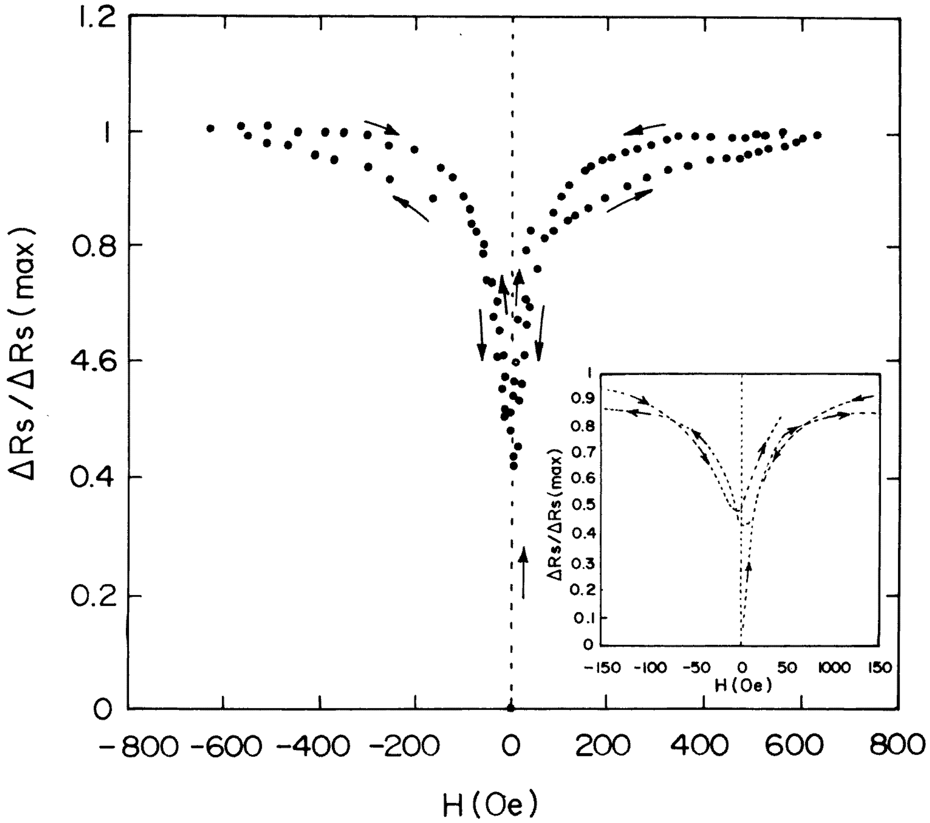

Figure 7. The surface resistance hysteresis curve of the BSCCO sample up to a maximum field of 630 Oe (at frequency 1.5 MHz); inset, an enlarged view.

The T_c of the YBCO–Ag sample was 90 K while that of the BSCCO sample was 80 K. The samples were cut in the shape of thin rods ($\sim$ 3 mm diameter, 1 cm length).

The r.f. surface resistance was measured using a Hewlett–Packard Q-meter. The samples were placed inside a coil which was connected to the Q-meter. The whole assembly was then dipped into liquid nitrogen and placed between the poles of an electromagnet. The axis of the coil was kept parallel to the magnetic field. Figure 3 shows the block diagram of the Q-meter. The surface resistance was measured according to the following procedure. Initially the shorting strip (S) was closed and the capacitor C tuned to obtain resonance. The values of capacitance (C_1) and quality factor (Q_1) were noted. This process was repeated after opening the shorting strip (S) and the values of capacitance (C_2) and quality factor (Q_2) were noted. The effective resistance (R_{sc}) and inductance (L_{sc}) of the sample and coil were calculated using the formulae

$$R_{sc} = (C_1Q_1 - C_2Q_2)/(\omega^2 C_1C_2Q_1Q_2)$$

$$L_{sc} = (C_1 - C_2)/(\omega^2 C_1C_2)$$

where ω is the frequency of the r.f. field. In all our measurements the value of Q_1 was 233.7 (determined by the Q-factor of the standard inductor L) and the value of Q_2 (determined by the Q-factor of L and Z in series) varied from 60 to 120.

The measured values R_{sc} (L_{sc}) are the sum of the coil resistance R_c (inductance L_c) and sample resistance R_s (inductance L_s). As only R_s and L_s are dependent on applied field

$$R_{sc} = R_c + R_s(H)$$

$$L_{sc} = L_c + L_s(H).$$

Only the changes of $R_s(H)$ and $L_s(H)$ with field are of interest, and we calculate the quantity

$$\Delta R_{sc}(H) = R_{sc}(H) - R_{sc}(H = 0) = \Delta R_s(H)$$

and

$$\Delta L_{sc}(H) = L_{sc}(H) - L_{sc}(H = 0) = \Delta L_s(H).$$

These values were normalized to $\Delta R_{s(max)}$ ($\Delta L_{s(max)}$) where $\Delta R_{s(max)}$ ($\Delta L_{s(max)}$) was the change in surface resistance (inductance) when the initial field was brought to its maximum value.

3. Results and discussion

The virgin curve and hysteresis of normalized surface resistance at frequency 1.5 MHz for the YBCO–Ag sample are shown in figure 4. The virgin curve initially increases rapidly with field, then shows a trend to saturate. The maximum applied field, 2 kOe, is much greater than H_{c1}

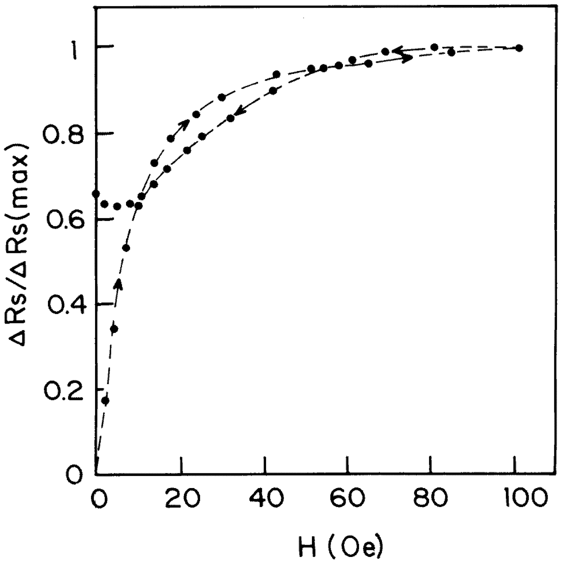

Figure 8. The surface resistance hysteresis curve of the BSCCO sample up to a maximum field of 100 Oe (at frequency 12 MHz).

in bulk samples and much less than H_{c2} [13] or H_g (the field at which the transition from vortex glass phase to vortex liquid phase occurs) [14] at 77 K. Our increasing virgin curve could not be fitted by an expression of the form $R_s = \alpha/(1 + H_0/H)$ where α and H_0 are constants as was done by Ramachandran *et al* [15] at microwave frequency. Surface resistance in decreasing field remains lower than that in increasing field, reaching a minimum at 30 Oe (figure 4). This behaviour agrees with the predictions of the two-level critical state model.

Results on BSCCO samples (figures 5–8) show more interesting features. The maximum applied field is much less than the H_{c2} of the grain, assuming that H_{c2} of the grain is of the same order of magnitude as that observed in single crystals [16]. However, as we shall see, we have here two different regimes in the surface resistance hysteresis curve depending on whether the maximum applied field is greater or less than the H_{c2} of the grain boundary.

Figure 5 shows the virgin curve (at 1.5 MHz) for surface resistance and inductance. The two curves almost overlap, indicating that pinning is weak [5].

Figure 6 shows the hysteresis of surface resistance when the maximum applied field is 40 Oe. Here the nature of the hysteresis curve is similar to the one obtained from the two-level critical state model. However when the maximum field is increased to 630 Oe the hysteresis curve follows a different behaviour (figure 7). As the field is decreased from its maximum value the surface resistance is initially higher than that in increasing field, resembling the one-level critical state. However, at a field of about 67.5 Oe, it crosses the curve for increasing field. This can be explained as follows. Below 67.5 Oe the sample consists of strongly superconducting grains separated by weakly superconducting grain boundaries. This is the situation in which the two-level critical state model is applicable. However, above 67.5 Oe the weakly superconducting grain boundaries become normal, and the sample consists of superconducting grains separated by normal metal. In this region the hysteresis in surface resistance comes only from the isolated grains. (The surface resistance of normal grain boundaries is reversible with respect to field.) This is a situation where the one-level critical state applies. Thus it is only below 67.5 Oe when intergranular regions become superconducting that the transition to two-level critical state behaviour occurs.

When the hysteresis measurement is made at 12 MHz (figure 8) the cross-over occurs at a lower value of field (~ 55 Oe). This cannot be explained on the basis of the ordered model we have been considering so far.

To explain this we note that the r.f. field penetrates only up to a finite thickness (penetration layer) within the sample. Thus in order to see the two-level critical state the r.f. field must penetrate over a sufficient length covering many grains and grain boundaries inside the sample. It has been argued by Ji *et al* [1] that in this case at high d.c. field several grains join together and act as one cluster. As long as the field penetrates only up to a few clusters the one-level critical state will persist. It seems reasonable to assume that this cluster size will decrease as we decrease the field. As the r.f. field at 12 MHz penetrates into a much smaller thickness than the r.f. field at 1.5 MHz the cross-over to the two-level critical state will occur at a lower value of field where these clusters become small enough for the r.f. field to penetrate over a large number of clusters. However a systematic study of this cross-over as a function of frequency is needed to confirm this.

4. Conclusion

The one-level and two-level critical state models successfully explain all the features observed in the surface resistance hysteresis of YBCO–Ag and BSCCO compounds. The hysteresis behaviour in BSCCO has been explained on the basis of a cross-over from a one-level to two-level critical state. A qualitative explanation of the frequency dependence of this cross-over point is given.

Acknowledgment

The author wishes to thank Professor S Ghatak for suggesting the problem and providing constant guidance.

References

[1] Ji L, Rzchowski M S, Anand N and Tinkham M 1993 *Phys. Rev.* B **47** 470
[2] Ji L, Rzchowski M S and Tinkham M 1989 *Phys. Rev.* B **39** 6607
[3] Pakulis E J and Osada T 1988 *Phys. Rev.* B **37** 5940
[4] Gittleman J I and Rosenblum B 1966 *Phys. Rev. Lett.* **16** 734
[5] Coffey M W and Clem J R 1991 *IEEE Trans. Magn.* **MAG-27** 2136
[6] Bardeen J and Stephen M J 1965 *Phys. Rev.* **140** A1197
[7] Bean C P 1962 *Phys. Rev. Lett.* **8** 250
[8] Anderson P W and Kim Y B 1964 *Rev. Mod. Phys.* **36** 39
[9] Halbritter J 1990 *J. Appl. Phys.* **68** 6315

[10] Zhen-peng S, Yong Z, Zu-Yao C, Xian-hui C and Qi-Yui Z 1989 *Solid State Commun.* **69** 1067

[11] Kao Y H *et al* 1990 *J. Appl. Phys.* **67** 353

[12] Sen D, Bhattacharya D, Ghatak S K and Chopra K L 1991 *Bull. Mater. Sci.* **14** 927

[13] Daumling M 1991 *Physica* C **183** 293

[14] Koch R H, Foglietti V, Gallagher W J, Koren G, Gupta A and Fisher M P A 1989 *Phys. Rev. Lett.* **63** 1551

[15] Ramachandran J S, Lofland S E, Bhagat S M, Manheimer M A and Tyagi S 1995 *Solid State Commun.* **93** 671

[16] Daumling M and Chandrashekhar G V 1992 *Phys. Rev.* B **46** 6422