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Possibility of striations in HTSC films caused by self-destabilization of the growth temperature

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Abstract

A new phenomenon of self-destabilization of the temperature of the growing high-Tc superconducting film is described and as a result, at least one of the reasons is found for the deterioration of superconducting parameters with increasing film thickness.

1. Introduction

One of the most important factors limiting the possibility to obtain high-T_c superconducting (HTSC) films with a high absolute value of the critical current is the deterioration of superconducting and microstructural parameters when the film thickness exceeds ~ 500 nm. The origin of this HTSC film behavior has not been explained up to now. One should assume, however, that the change in properties with increasing film thickness points directly to the unstable character of film growth conditions. This paper deals with one of the causes of such instability, namely, the temperature self-destabilization.

Much effort has been made to provide constant temperature conditions for HTSC film growth [1-4]. It is usually expected that providing a reliable thermal contact between the substrate and the heater is sufficient to keep the temperature constant at the growth surface [1-3]. This, however, does not take into account heat transfer resulting from heat irradiation. This mechanism should be important for conventional growth temperatures (~ 800°C) for HTSC films, and simultaneously it should depend considerably on IR film parameters and lead to a deviation of heat balance during film growth.

The present investigation studies the process of temperature self-destabilization in growing HTSC films as well as the possibility of specific non-uniformity formation (striations) due to such fluctuations of the growth temperature.

2. Temperature equation for growing the film

Let us consider the heat transfer in the film F - substrate S system when the substrate is in ideal contact with a metallic surface M of constant temperature T_m (see Fig. 1). Let us also assume that the substrate is transparent for the given spectrum of the thermal radiation, though this condition is usually only partially fulfilled. Both the film F at temperature T and the metallic surface M serve as sources of the thermal radiation. The film, representing an effective semitransparent thermal screen, partly reflecting radiation, creates a kind of trap for the thermal radiation 1 (see Fig. 1), which as a result of multiple re-reflections between the film and the surface M escapes through the film surface only in a reduced form (flux 2, Fig. 1), while the film is still transparent. Thermal fluences arising due to "interior" and "exterior" irradiation $\dot{q}_1$ and $\dot{q}_2$, respectively can be calculated from the following equations obtained in a conventional manner:

$$\dot{q}_1 = [E_m(1-R') - E\,A_m]/(1 - R'R_m)\,, \qquad (1)$$

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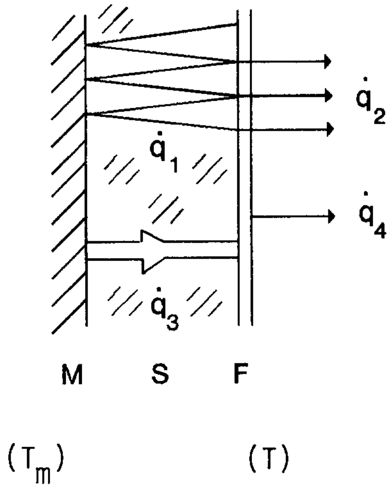


Figure 1. Scheme of heat transfer.
M - heater surface, S- substrate, F - film.

$$\dot{q}_2 = (E_m\tau + E'R_m\tau)/(1 - R'R_m) , \quad (2)$$

where $E_m = A_m \sigma T_m^4$, $E' = A' \sigma T^4$. σ denotes the Stefan-Boltzmann constant and R', τ, A' are respectively the IR energy coefficients of reflection, transmission and absorption averaged over the thermal radiation spectrum, using a weight function corresponding to the spectral distribution of this irradiation (prime indicates a situation where the irradiation enters the film from the substrate side). R_m, A_m denote analogous coefficients of reflection and absorption for the boundary: metallic surface - substrate. These coefficients are connected with each other through the obvious equations $R' + \tau + A'= 1$, $R_m + A_m= 1$.

Heat transfer from the heater to the film is affected not only by radiation but also by ordinary thermal conductivity which causes a thermal flux $\dot{q}_3 = (\lambda / l)(T_m- T)$, where the thermal conductivity coefficient λ and the length l may generally have effective values that take into account not only the thermal conductivity of the substrate but also the thermal conductivity of the contact interlayer between substrate and heater.

If the thermal losses $\dot{q}_4 = A \sigma T^4$ resulting from the thermal radiation of the film itself are also considered, one obtains from the heat balance condition:

$$\dot{q}_1 - \dot{q}_2 + \dot{q}_3 - \dot{q}_4 = 0 \quad (3)$$

the following equation for the film temperature T as a function of the film thickness t:

$$a(t)\, T^4 + b(t)\, T - c(t) = 0, \quad (4)$$

where

$$a(t) = \sigma A'(t)[A_m + R_m\tau(t)] + \sigma A(t)[1 - R_mR'(t)],$$

$$b(t) = (\lambda / l)[1 - R_mR'(t)],$$

$$c(t) = \sigma A_mT_m^4A'(t) + (\lambda / l)[1-R_m R'(t)]\, T_m.$$

It is quite obvious that in order to use this equation for the calculations of T(t), it is necessary to determine the thickness dependences of the optical parameters R, τ and A.

3. Measurements of the optical parameters

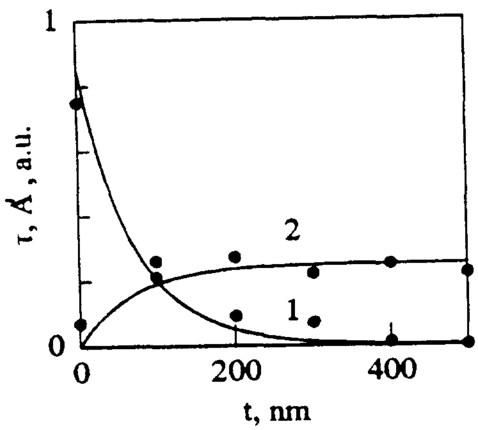
Measurements of the dependence of transmission and absorption coefficients on the varying film thickness were performed by a calorimetric method using the thermal radiation of the heater itself, heated at T_m = 850°C. During the measurements the samples were placed parallel to the heater surface at a distance of about 5 cm. The coefficients of absorption A, A′ were determined as the relation of the heating rate of the $SrTiO_3$ substrate with an $YBa_2Cu_3O_{7-x}$ film to the heating rate of the analogous substrate covered with a "black" carbon layer absorbing practically 100 % of the incident radiation. The transmission coefficient of the film was derived from the relation between the heating rates of the substrate covered with a "black" layer which was either screened or not screened. The preliminary measurements showed that the absorption coefficient of the clean substrate is about 0.08. This value as well as the reflection coefficient R_S of the clean substrate surface were employed for the final calculation of the measured parameters. Reflection coefficients R′ were calculated according to $R'= 1 - A'- \tau$.

The results of the measurements of the optical parameters of a sample series obtained by laser ablation versus film thickness are shown in Fig. 2 (dots). The experimental data shown in this figure can be approximated by the following equations:

$$A' = A_i [1 - \exp(-t / t_0)], \quad (5)$$

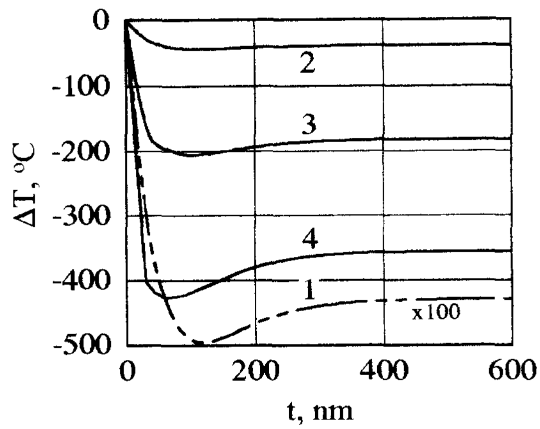
$$\tau = (1 - R_S) \exp(-t / t_0), \quad (6)$$

where t_0 is the average penetration depth of the IR radiation in the film and A_i is the absorption



t, nm

Figure 2. Experimental thickness dependence of IR coefficients of transmission τ (1) and absorption A' (2) for $YBa_2Cu_3O_{7-x}$ films grown on $SrTiO_3$.



t, nm

Figure 3. Dependences of $\Delta T = T - T_m$ on film thickness. The curves 1-4 correspond to $\lambda = 5$, 0.5, 0.05, 0,005 W/m K; $T_m = 850°C$, $l = 0.5$ mm, $R_S = 0.14$.

coefficient of a film with a thickness $t >> t_0$. Fig. 2 demonstrates that equations (5) and (6) reasonably well describe the experimentally obtained thickness dependences at $t_0 = 70$ nm and $A_i = 0.25$. These dependences can be directly used to calculate the desired temperature variation of the film versus its thickness during deposition.

4. Results of calculation and discussion

The calculations of the temperature deviations $\Delta T(t) = T - T_m$ in the film during its growth are shown in Fig. 3 for various values of the thermal conductivity coefficient λ. The calculation was performed by using equation (4) as well as formulas (5) and (6). The IR absorption coefficient was approximated by $A_m \approx 0.05$. This corresponds to the value for a silver paste, which is usually employed to provide the thermal contact to the substrate [2]. Curve 1 illustrates the case of a practically ideal thermal contact between the substrate and the heater. As the film grows, the temperature T decreases by about 5°C, reaches its minimum at t = 130 nm, and increases again by about 0.8°C.

The deterioration of the thermal contact between the substrate and the heater leads to a pronounced overcooling of the film by tens or even hundreds of degrees when the film reaches the thickness of 50-100 nm (see curves 2-4 in Fig. 3). It may be seen from the plots of Fig. 3 that overcooling is accompanied by the displacement of the temperature minimum to smaller thicknesses.

The analysis of the calculated dependences (4)-(6) shows that this temperature minimum originates from the competition between the increasing irradiation losses of the film with growing thickness (flux $\dot{q}_4$) and the increasing efficiency of the irradiation trap formed between the film and the heater surface.

Thus we can conclude that a constant temperature of the heater does not lead to temperature stabilization of the growth surface. The resulting self-destabilization of the film temperature should lead to the formation within the film of HTSC layers with improved and deteriorated properties (some kind of striations), because the film quality is generally sensitive to changes of the growth temperature by more than ~ 2°C. If the optimal temperature of crystallization T_O is provided at the initial stage of growth, the growth of the rest of the film would be subjected to overcooling. The film would then consist of a thin HTSC interlayer of good quality covered with an overlayer with deteriorated parameters. Likewise, when $T_{min} < T_O < T_m$, striated films shuold be formed, which consist of multiple layers with deteriorated and non-deteriorated properties.

The experimental evidence of such a behavior was obtained from the measurement of the superconducting transition temperature for a series of $YBa_2Cu_3O_{7-x}$ films with various thicknesses. It was found that at $T_m = 835°C$ the formation of an interlayer with non-deteriorated HTSC properties starts at a critical thickness $t_C \approx 100$ nm. If the temperature T_m is reduced to 825°C then t_C decreases to 40 nm. In both cases this is accompanied by a saturation of the critical current when the thickness exceeds ~ 1.5 t_C. This obviously points to the appearance of layer-type non-uniformities of the growing film which should result from the above discussed self-destabilization model. Moreover this model might explain the existence of an initially deteriorated layer near the substrate - film interface which might not be caused by interdiffusion.

It is important to note that in general the appearance of even one striation during film growth may lead to an irreversible deterioration of the further film growth. Therefore, the main problem of HTSC film growth is found in providing constant growth temperature at the initial stage of the film deposition. It seems to be quite difficult to meet such a requirement because of the rapid change of the thermal balance due to a rapid increase of the IR emission at this stage of growth.

The present results show that the worst conditions for film growth occur when the thermal contact of the substrate with the heater is poor, especially in the case of heating a "free" substrate with optical or thermal radiation [5]. Nevertheless, even in the case of a perfect thermal contact the change of the film temperature has to be taken into account, which must be compensated by a respective change of the heater temperature. The possibility of using a pyrometer control of the substrate temperature [1] for this purpose seems to be questionable because in this case one is always confronted with the necessity to subdivide the total radiation flux in contributions from the substrate and the growing film. In general this separation cannot be usually done in a precise manner.

Thus it seems obvious that to provide constant temperature conditions for HTSC film deposition special methods should be developed which would permit to eliminate or at least to decrease the influence of a thermal irradiation change on the film temperature.

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