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КОМПЬЮТЕРНЫЕ ТЕХНОЛОГИИ В ФИЗИКЕ

THEORETICAL EVIDENCES FOR SUPERHEATING DURING TRACK FORMATION IN HIGH-T_c SUPERCONDUCTORS

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At present the thermal spike description of heavy ion track formation in high-T_c superconductors is confronted with serious difficulties caused by a high sensitivity of theoretical track radii to a small change of the electron diffusivity value. This sensitivity is caused by a bifurcation point found numerically in the model and resembles the impossibility of the classical gas description in the mechanics framework due to a dramatic dependence of particle trajectories on small variations of the initial conditions. For solution of the problem, we suggest a thermal explosion model which takes into account such a nonequilibrium process as superheating known from laser-induced phase transformations in solids. This allows us to essentially stabilize the theory. For the first time we give a quantitative description of tracks in $YBa_2Cu_3O_{7-x}$ with both elliptical and circular cross sections.

Описание процессов трекообразования в высокотемпературных сверхпроводниках в рамках модели термопика в настоящее время столкнулось с серьезными трудностями, связанными с высокой чувствительностью теоретических радиусов треков к малым изменениям электронной температуропроводности. Столь высокая чувствительность обусловлена чисто математическими особенностями модели — бифуркацией, обнаруженной в ходе численных экспериментов. Неустойчивость такого типа напоминает невозможность долгосрочного предсказания траекторий частиц газа методами классической механики из-за высокой чувствительности предсказаний к неопределенности начальных условий. В настоящей работе для решения данной проблемы предлагается модификация модели термопика (модель теплового взрыва), учитывающая неравновесные процессы перегрева при плавлении. Подобные эффекты были обнаружены ранее в экспериментах с короткими лазерными импульсами, вызывавшими фазовые переходы в твердых телах. Учет этих процессов позволяет существенно улучшить предсказательную способность теории. Впервые дается количественное описание размеров треков в $YBa_2Cu_3O_{7-x}$ как кругового, так и эллиптического типа.

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INTRODUCTION

The first attempt to explain track formation in high-T_c superconductors appealed to the ionic spike model [1] based on simple and clear picture of the electrostatic «explosion» of a positive charged region supposed to develop in the wake of the «bombarding» ion [2]. Lately it has been realized that the same experimental data can be qualitatively interpreted as well in the context of the thermal spike model [3] considering track formation to be

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the result of melting and subsequent quick solidification of the material found itself in the immediate vicinity of the passing ion [4]. Physical reasons for such a suggestion were simple and convincing too. Firstly, nanodiffraction from the region of tracks in $\mathrm{YBa_2Cu_3O_{7-x}}$ showed that it is amorphous as if it was melted indeed and afterward quenched, memorizing in that way a disorder accompaning the melting. Secondly, the experiments showed that the amorphous region had a distinct edge in the surrounding monocrystal, which was not so natural from the ionic spike model point of view. Thirdly, transmission electron microscopy revealed lattice distortion corresponding to dilation of the material inside tracks [4, 5], and this fact can be also interpreted as the result of melting accompanied with expansion of the material.

The first attempts to apply the thermal spike model (TSM) to description of track formation in high-T_c superconductors were oversimplified and neglected some important information about materials. For example, disregarding the latent heat of melting resulted in prediction of track radii to be greater than the experimental ones. To justify this difference, an interesting hypothesis of «epitaxial regrowth» was suggested according to which the molten region does not all become amorphous, but the outer part of it should undergo recrystallization [4]. Now this assumption looks premature and evidently incorrect [6]. In [7], a phenomenological approach based on the thermal spike concept was proposed to explain the evolution of track sizes with energy deposition for irradiated $\mathrm{YBa_2Cu_3O_{7-x}}$ and $\mathrm{Bi_2Sr_2CaCu_2O_8}$ superconductors. Although this useful model was successful in its design, it contained some parameters independent of the physical properties of the materials and could be only considered as a preliminary investigation of the problem.

A more detailed model of track formation in $\mathrm{YBa_2Cu_3O_{7-x}}$ based on a system of coupled equations for electron and atom temperatures was proposed in [8] by analogy with TSM developed in Caen [9] for description of latent track formation in amorphous metals and semiconductors. The mean free path of electron scattering, $\lambda = \sqrt{D_e \tau}$, was assumed to be the only free parameter in this approach. Here D_e is the diffusivity of the excited electrons in the vicinity of ion trajectory which, for a given material, is usually supposed to be a constant belonging to the range of 1–2 cm^2/s [11]. Parameter τ is the electron–atom relaxation time approximately determined in femtosecond laser experiments [12, 13]. Other quantities used in the model are known macroscopic characteristics of an irradiated matter such as thermal conductivities of electrons and atoms, K_e and K_i, their specific heats, C_e and C_i, density ρ of solid and liquid phases, melting temperature T_m, and heat of fusion Q_f. The value of parameter $\lambda \simeq 18$ nm found in [8] for $\mathrm{YBa_2Cu_3O_{7-x}}$ was close to the corresponding magnitude obtained for amorphous metals and semiconductors, electron–atom relaxation time τ turned out to be in a good agreement with femtosecond laser experiments and all that seemed to be quite reasonable. However, simple analytical estimations fulfilled in [8] have shown that the experimentally observed dependence of track radii on energy deposition can be explained only in the case if one takes into account an approximate linear dependence of τ on T_e, and such a dependence can be justified in the Allen theory framework [14]. Thus, at this point the description of track formation in $\mathrm{YBa_2Cu_3O_{7-x}}$ displayed a necessity to step aside from the Caen version of TSM, in which τ is supposed to be temperature-independent.

Further study of the model revealed the following surprising peculiarities. Firstly, the value of D_e was *found*, instead of a theoretically motivated *assumption* in the Caen TSM, to be approximately equal to 1 cm^2/s from the requirement that the model should describe the experimental track radii, and *no other free parameters* were used at all [6]. Thus, prospects to formulate the quantitative theory of track formation in high-T_c superconductors appeared at the horizon. At the same time, very high sensitivity of track radii to a small ($\sim 0.1\%$) change of D_e was established [6]. Such a precision is actually unachievable neither experimentally nor theoretically and this renders the theory utilization and verification almost impossible.

In the present paper, we propose a solution to the problem taking into account the superheating process known from laser-induced phase transformations in solids [15]. This allows us to stabilize the solutions to some reasonable extent. Besides, we complete the description of the available experimental data for $YBa_2Cu_3O_{7-x}$ considering not only tracks with the circular section (arising in the case when the ion trajectory is perpendicular to the superconducting, a–b, plate), but also tracks with the elliptical section. Since a comprehensive description of the model can be found elsewhere [6], here we give only a brief outline of the theory focusing on its appropriate modifications. Results of the mathematical simulation and their comparison with experimental data are given at the end.

1. DESCRIPTION OF THE MODEL

We assume the following system of two coupled nonlinear differential equations for electron and atom temperatures, T_e and T_i, respectively:

$$\rho C_e(T_e)\frac{\partial T_e}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left[rK_e(T_e)\frac{\partial T_e}{\partial r}\right] + \frac{1}{r^2}\frac{\partial}{\partial \varphi}\left[K_e(T_e)\frac{\partial T_e}{\partial \varphi}\right] - g(T_e - T_i) + q(r,\varphi,t), \quad (1)$$

$$\rho C_i(T_i)\frac{\partial T_i}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left[rK_i(T_i)\frac{\partial T_i}{\partial r}\right] + \frac{1}{r^2}\frac{\partial}{\partial \varphi}\left[K_i(T_i)\frac{\partial T_i}{\partial \varphi}\right] + g(T_e - T_i), \quad (2)$$

where the incident ion is supposed to be parallel to z and $r^2 = x^2 + y^2$. When the ion trajectory is directed along c axis, one can ignore the φ dependence and the system (1), (2) is reduced to that used in [6] for description of tracks with circular cross sections.

When the incident ion is parallel to the [100] or [010] directions the defects appear elliptical in shape [4]. We shall try to explain this fact by an anisotropy of the thermal conductivity of $YBa_2Cu_3O_{7-x}$ in the a–c and b–c planes. The total in-plane, K_{ab}, and out-of-plane, K_c, thermal conductivity were experimentally studied in [16–18]. Whereas a ratio of these values extracted from [17, 18] is

$$\frac{K_{ab}}{K_c} \approx 17, \quad (3)$$

the paper [16] gives the value, $K_{ab}/K_c \approx 7\pm^{4}_{2}$. The first result seems to be more convincing since according to [18] the out-of plane thermal conductivity obtained in [16] was overestimated because of inhomogeneous distribution of heat current. Authors of [18] also infer that their data, measured between the room temperatures and 70 K, agree to a phonon-dominated out-of-plane thermal conductivity. One can adjust this conclusion with the ratio (3) assigning to the out-of-plane thermal diffusivity the value

$$D_{i,c} \approx \frac{D_{i,ab}}{7.5}, \quad (4)$$

where $D_{i,ab} \approx 1.81 \cdot 10^{-2}$ cm^2/s is in-plane thermal diffusivity of $YBa_2Cu_3O_{7-x}$ at the room temperature [6]. The final formula for the out-of-plane thermal diffusivity can be written in the form:

$$D_i(\varphi) = \sqrt{D_{i,ab}^2 \cos^2 \varphi + D_{i,c}^2 \sin^2 \varphi}.$$

According to [18] at temperatures $T < 300$ K phonons contribute also a great deal to the in-plane thermal conductivity, K_{ab}, of $YBa_2Cu_3O_{7-x}$. In fact, the dominant lattice contribution of phonons to K_{ab} was established already in early experiments and was confirmed in subsequent investigations (see review [19]). As in our previous paper [6], a relative contribution of free carriers to the total thermal conductivity in the normal state of $YBa_2Cu_3O_{7-x}$ is taken from [19, p. 215]: $K_{e,ab}/K_{ab} = 0.31$. The value of $D_{e,ab}$ at the room temperatures is estimated using $K_{e,ab}$ and a trustworthy experimental value of the electronic thermal capacity [6].

A separate issue is a behaviour of out-of-plane thermal conductivity at high temperatures where charge excitations should play an essential role. The simplest parametrization describing such a behaviour *and consistent with TSM* is the following: $D_{e,c} \approx 0$ for $T_e < T_{\rm th}$ and $D_{e,c} = D_{e,ab}$ for $T_e \geqslant T_{\rm th}$, where $T_{\rm th}$ is some threshold temperature considering as a parameter of the model. Here, the value of the in-plane thermal diffusivity, $D_{e,ab}$, can be fixed at the stage of description of the circular tracks arising in the same material at the same energy of the incident ions, but at another orientation of the specimen.

2. THERMAL EXPLOSION MODEL (TEM)

According to TSM, all primary energy losses of the incident ion are concentrated in the electron subsystem, and the further electron–atom energy transfer is accompanied by the electron and atom heat conduction (see Eqs. (1) and (2)). Since at high temperatures $T_e \geqslant 10^3$ K, the value of K_e in $YBa_2Cu_3O_{7-x}$ is at least an order greater than K_i, the initial electron hot spot passes far ahead of the phonon one and has time to spread over a considerable part of the track cross section already at the early stage of the electron–atom relaxation process. This leads to beginning of an almost synchronous volumetric electron–atom energy transfer. The synchronization of the process is also enhanced by a specific dependence of the electron–atom coupling, g, on the electron temperature, T_e, in the material: the higher T_e is, the smaller value of g remains [6]. Such a nature of atom heating should result in the melting process very different from melting near equilibrium.

Indeed, phase transition near equilibrium point can be described by the nucleation theory according to which the melt originates from a small spherical cluster of atoms which lost their structural order. Beyond a critical size, the cluster will expand throughout the volume until the entire specimen is transformed into the liquid phase (before a sizeable amount of heat has time to come from outside). There is a suitable analogy of this. Let us consider a train moving from a higher platform to a little lower one separated from the first platform by a narrow hill. Here a difference of potential energies of a railway-carriage on two different platforms corresponds to the difference of free energies in the solid and liquid phases, per unit of volume or mass, and the hill symbolizes a free energy of the interface. According to the analogy, the quantity of atoms in the melt is described by a number of carriages got over the narrow hill. It is worth noting that only a small part of the train should be lifted at each instant of time to transfer it into the energy preferable state. (A resembling method of transferring his body over the bar is used by a skilled high jumper.) The picture changes drastically when a powerful electron–phonon energy transfer takes place almost simultaneously in an essential part of the volume. Now the «train» is blown up to the height of the «hill», and perhaps higher, due to a powerful shock from the outside (just as a vigorous high jumper should not necessarily repeat all movements of a feeble one to clear the same obstacle).

Definite traits of superheating were revealed experimentally in laser-induced melting of thin films [15]. In particular, it was found that the energy required for the rapid melt metamorphosis exceeds the equilibrium melting heat in Al by a factor of 1.4–2.6, depending on the rate of transformations which the authors were able to realize. The times of nonequilibrium phase transitions observed in [15] were located in the range from 10 to 1000 ps, which were even longer than the typical times of keeping the melting conditions in the tracks, $t \sim 1{-}10$ ps [6]. Therefore, the idea of superheating during track formation seems to be quite natural. We assume that the change of the atom temperature under the superheating conditions is evaluated with the formula $\Delta T_i \approx \Delta Q/C_i$, where ΔQ is an increase of the lattice energy per unit of mass, and C_i is taken at the room temperature. The only free parameter of our thermal explosion description is the temperature of superheating, $T_{\rm sh}$, which describes a minimum value of atom temperature that should be mounted for destroying a structural order of the substance.

3. COMPARISON WITH THE EXPERIMENT

A numerical solution of system (1), (2) was assisted with a finite-difference scheme due to Samarskii (see Appendix).

Experimentally observed radii of tracks, $r_{\rm exp}$, in $\rm YBa_2Cu_3O_{7-x}$ single crystal with [001] axis oriented parallel to the incident ion beam are given in the table along with results of our calculations. As compared with [6], the sensitivity of track radii to small changes of the value of electron thermal diffusivity goes down to one order: one can see that setting the precision of D_e over the range from 2 to 12% turns out to be quite enough to fit the theoretical track radii.

Experimentally observed radii of tracks, $r_{\rm exp}$, in $\rm YBa_2Cu_3O_{7-x}$ single crystal taken from [20] and results of their theoretical description on the base of TEM. Energy deposition dE/dx was calculated using [21]. Pseudo-diffusivity of electrons, $D_e \equiv K_e/\rho C_e$, was taken to adjust the theoretical track radii to $r_{\rm exp}$

Ion	Energy, MeV/amu	dE/dx, keV/nm	$r_{\rm exp}$, nm	$r_{\rm th}$, nm	D_e, cm^2/s
^{129}Xe	1.3	26.2	2–3	2.8–2.3	0.15–0.18
^{129}Xe	2.6	30	2.5	2.5	0.18
^{129}Xe	10	27.9	1.3	1.4	0.17
^{129}Xe	27	18.7	1.3	1.3	0.08
^{129}Xe	41	14.8	0.56	0.7	0.055
^{208}Pb	3.7	43.7	4	4.0	0.2
^{208}Pb	10	42.5	3	3.0	0.23
^{208}Pb	20	37	3.5	3.5	0.14
^{208}Pb	25	34.5	3	3.0	0.14
^{197}Au	1.52	36.2	3.8	3.7	0.18

Dependence of obtained electron pseudo-diffusivity D_e on the energy deposition dE/dx is shown in Fig. 1. Figure 1 shows that parameter D_e for $YBa_2Cu_3O_{7-x}$ cannot be considered independent of the electron temperature, as it is supposed in the Caen version of TSM. This conclusion is also supported marginally by Fig. 2, where D_e is shown as a function of electron temperature.

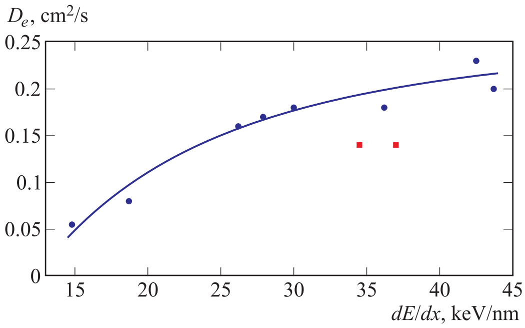

Fig. 1. Dependence of electron pseudo-diffusivity, D_e, on energy deposition in $YBa_2Cu_3O_{7-x}$ found using the thermal explosion model (points). The solid line presents the smoothing

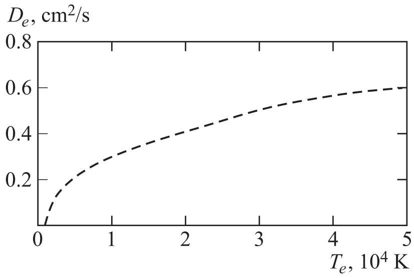

Fig. 2. Theoretical $D_e(T_e)$ dependence for amorphous carbon extracted from [22]

High-resolution images of 300-MeV Au^{24+}-irradiated $YBa_2Cu_3O_7$ thin films for two cases, when the incident ion is parallel to the [100] and [001] directions, were obtained in [23]. Using these data one can estimate sizes of tracks with circular and elliptical cross sections: $r \approx 3.8$ nm, and $r_1 \approx 6.3$, $r_2 \approx 4.7$ nm, accordingly. Our preliminary estimations in the frame of TEM give for circular tracks $r \approx 3.7$ nm, and for elliptical ones $r_1 \approx 5.5$, $r_2 \approx 4.3$ nm. Parameters $T_{\rm th}$, and $T_{\rm sh}$ were taken $\approx 1.1 \cdot 10^4$ K and $4T_m$, accordingly.

CONCLUSION

At present the thermal spike picture of track formation in $YBa_2Cu_3O_{7-x}$ high-T_c superconductor in a consistent way embraces all available experimental data concerning the properties of the substance [6]. This makes the model credible at least in general outline. Thus, one can conclude that the amorphous state of the track region, observed in nanodiffraction experiments, is, almost certainly, a result of melting and subsequent quick solidification. However, our theoretical estimate makes it clear that a lifetime of conditions favourable for the liquid phase existence in the track is too short to provide usual equilibrium melting. Similar processes were studied experimentally during laser-induced melting in thin films [15], and the principal inference is that under those conditions the substance should be exposed to superheating. Numerical estimations performed in the present paper have shown that superheating in $YBa_2Cu_3O_{7-x}$ can occur only when the value of electron diffusivity at high temperatures, D_e, is less than it is often assumed in TSM. Actually, it should be even less than in-plane thermal diffusivity of electons at room temperatures, $D_{e,ab} \simeq 0.26-0.52$ cm^2/s [6]. Established dependence of D_e on energy deposition, or on T_e, means that the suggestion of the Caen version TSM about the brownian character of transportation of electrons is inapplicable for $YBa_2Cu_3O_{7-x}$. To describe this value, one should use a more detailed kinetic model taking into account a dependence of the concentration of charged carriers and their scattering cross sections on T_e. The problem of high sensitivity of theoretical track radii to small changes of D_e, posed in [6], can be relaxed appreciably in the TEM framework. The fact is that the decrease of D_e results in moving off a bifurcation point which is an immediate cause of the instability [6]. Radii of tracks with circular cross section lying in the superconducting plane can be described in the context of TEM quite satisfactorily. Sizes of elliptical tracks created in $YBa_2Cu_3O_{7-x}$ when incident beam is oriented along [100] direction can be explained in the frame of TEM as well, if one takes into account anisotropic thermal conductivity of the material.

APPENDIX

Numerical calculations are carried out on a grid of variables r, φ and t with spatial, angle and time steps h_r, h_φ and h_t, respectively

$$r_j = r_{\min} + jh_r, \quad j = 0, \ldots, n_r, \quad h_r = (r_{\max} - r_{\min})/n_r,$$

$$\varphi_k = kh_\varphi, \quad k = 0, \ldots, n_\varphi, \quad h_\varphi = \pi/2/n_\varphi,$$

$$t_l = lh_t, \quad l = 0, \ldots, n_t, \quad h_t = t_{\max}/n_t,$$

where $r \in [r_{\min}, r_{\max}]$, $\varphi \in [0, \pi/2]$, $t \in [0, t_{\max}]$, n_r, n_φ and n_t are numbers of partitions.

For numerical solution, the system (3), (4) was approximated by the following explicit finite-difference scheme [24]:

$$\rho C^{\,l}_{e\,j,k} \frac{T^{\,l+1}_{e\,j,k} - T^{\,l}_{e\,j,k}}{h_t} =$$

$$= \frac{1}{r_j}\mathbf{\Lambda_1}\left[r_j\, K^{\,l}_{e\,j,k}\, T^{\,l}_{e\,j,k}\right] + \frac{1}{r_j^2}\mathbf{\Lambda_2}\left[K^{\,l}_{e\,j,k}\, T^{\,l}_{e\,j,k}\right] - g^{l}_{j,k}(T^{\,l}_{e\,j,k} - T^{\,l}_{i\,j,k}) + q^{l}_{j,k}, \quad (5)$$

$$\rho C^{\,l}_{i\,j,k} \frac{T^{\,l+1}_{i\,j,k} - T^{\,l}_{i\,j,k}}{h_t} = \frac{1}{r_j}\mathbf{\Lambda_1}\left[r_j\, K^{\,l}_{i\,j,k}\, T^{\,l}_{i\,j,k}\right] + \frac{1}{r_j^2}\mathbf{\Lambda_2}\left[K^{\,l}_{i\,j,k}\, T^{\,l}_{i\,j,k}\right] + g^{l}_{j,k}(T^{\,l}_{e\,j,k} - T^{\,l}_{i\,j,k}), \quad (6)$$

where

$$\mathbf{\Lambda}_1(r_j K^l_{j,k} T^l_{j,k}) = \frac{1}{h_r}\left[r_{j+\frac{1}{2}} K^l_{j+\frac{1}{2},k} \frac{T^l_{j+1,k} - T^l_{j,k}}{h_r} - r_{j-\frac{1}{2}} K^l_{j-\frac{1}{2},k} \frac{T^l_{j,k} - T^l_{j-1,k}}{h_r}\right],$$

$$\mathbf{\Lambda}_2(K^l_{j,k} T^l_{j,k}) = \frac{1}{h_\varphi}\left[K^l_{j,k+\frac{1}{2}} \frac{T^l_{j,k+1} - T^l_{j,k}}{h_\varphi} - K^l_{j,k-\frac{1}{2}} \frac{T^l_{j,k} - T^l_{j,k-1}}{h_\varphi}\right],$$

$$T^l_{j,k} = T(r_j, \varphi_k, t_l),\ C^l_{j,k} = C(T^l_{j,k}),\ K^l_{j,k} = K(T^l_{j,k}),\ g^l_{j,k} = g(T^l_{j,k}),\ q^l_{j,k} = q(r_j, \varphi_k, t_l),$$

$$r_{j+\frac{1}{2}} = \frac{r_j + r_{j+1}}{2}, \quad K^l_{j+\frac{1}{2},k} = \frac{K(T^l_{j,k}) + K(T^l_{j+1,k})}{2}, \quad K^l_{j,k+\frac{1}{2}} = \frac{K(T^l_{j,k}) + K(T^l_{j,k+1})}{2}.$$

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