Formation of columnar defects in high-T_c superconductors by swift heavy ions

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Experimental data on the variation of the size of columnar defects with the electronic stopping power S_e are analyzed for Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O. A model is applied that is based on the thermal spike concept and which was previously successfully used for irradiated insulating materials. The width of the thermal spike $a(0)$ obtained from the analysis is 4.4 and 4.65 nm for Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O, respectively, which are in good agreement with the value for insulators. The fraction of the energy deposited to the thermal spike in the phonon system gS_e is lower in high-T_c superconductors (HTCS's) than in insulators. This reduction is attributed to the anisotropy of the electron scattering. The model is suitable to predict the size of columnar defects in HTCS's. [S0163-1829(96)03641-7]

I. INTRODUCTION

Recently, high-energy heavy ion irradiation has been applied by several groups to increase the critical current density j_c in high-T_c superconductors (HTCS's). The nonsuperconducting columnar defects created by the energetic ions were used to pin the flux lines (for a review see Ref. 1). Civale *et al.* found that in Y-Ba-Cu-O the pinning is most effective when the defects are continuous and the applied magnetic field is parallel to the axis of the columns.[2] Due to columnar defects j_c largely increases at all temperatures and fields. The optimum result is expected when the diameter of the continuous columnar defects is of the order of the coherence length. Therefore, it is of primary importance to predict the range of ion-beam parameters which is suitable for the creation of columnar defects with the optimum size.

High-resolution electron microscopy (HREM) studies revealed that the tracks along the trajectory of energetic ions are amorphous. It is well known from the systematic studies reported in Ref. 1 that in yttrium iron garnet (YIG) continuous tracks are formed only at $S_e > S_{eo}$ when the effective radius of the track R_e exceeds a certain R_{eo} value. Below this size the defects consist of beads of small spheres and elongated defects. In a three-dimensional anisotropic superconductor like Y-Ba-Cu-O, these defects are less effective for flux-line pinning than long columnar defects. Therefore, it is important that ion beams with $S_e > S_{eo}$ should be used for flux-pinning irradiation.

An attempt has been already made by Tombrello[3] to describe track evolution in Y-Ba-Cu-O. The estimation is based on the expression which was obtained previously by Tombrello for the secondary electron energy density. A reasonable agreement was found between the measured and the predicted track sizes. It was also noted that higher energy density is required in Y-Ba-Cu-O than in YIG to create tracks.

In this paper a different approach is applied. Recently, a model was proposed by the author for the interpretation of experiments on track formation in insulating materials.[4] The analysis revealed that track formation proceeds identically in quite different materials (YIG, ferrites, mica) if thermal properties are properly taken into account. In the present paper this model is applied to HTCS's. It is shown that along with the similarities there are also important differences between insulators and HTCS's in respect to track formation.

The paper is structured as follows: in Sec. II we give a brief outline of the model, the experimental data are reviewed in Sec. III, in Sec. IV we interpret the results according to the model, and, finally we draw the appropriate conclusions in Sec. V.

II. THE MODEL

It is assumed that a high-temperature region, a thermal spike is formed along the trajectory of an energetic ion. If the temperature exceeds the melting point T_m the material melts within a cylinder. As a result of the high cooling rate ($\approx 10^{14}$ K/s) an amorphous phase can be formed. The model is based on two assumptions: (i) the radial distribution of the temperature increase in the phonon system is a Gaussian function with an initial width $a(0)$, (ii) the radius of the melted (R_o) and the experimentally determined amorphized cylindrical volumes (R_e) are equal. In the approximation applied in Ref. 4 the model provides the following equations for R_o:

$$R_o^2 = a^2(0)\ln(S_e/S_{et}), \quad 2.7 \geqslant S_e/S_{et} \geqslant 1, \tag{1}$$

$$R_o^2 = [a^2(0)/2.7](S_e/S_{et}), \quad S_e/S_{et} \geqslant 2.7, \tag{2}$$

$$S_{et} = \rho \pi c a^2(0) T_o / g, \tag{3}$$

where gS_e is the fraction of S_e deposited in the thermal spike, c, ρ, and T_o are the average specific heat, the density, and the difference between the melting T_m and irradiation temperatures T_{ir}, respectively. The $a(0)$ parameter characterizes the width of the temperature distribution and g is the efficiency of the energy deposition in the thermal spike (for details see Ref. 4).

The model was successfully applied to the explanation of the variation of damage cross section with S_e in insulating materials. It was shown that both $a(0)$ and g have equal values (within experimental error) for a number of insulators.[4,5] The comparison with experiments proved the validity of assumptions (i) and (ii). The damage cross-section velocity effect reported by Meftah *et al.*[6] was explained based on the model.[7] A quantitative agreement was also found between the experiments and the predictions of the model in respect of the temperature dependence of track formation.[8] Equations (1)–(3) require that the physical parameters of the target affect track formation through the term $\rho c T_o$. Comparison with experiments confirm this consequence of the model, as well.[4,8]

III. EXPERIMENTAL DATA

Different experimental methods were applied for the measurement of the track sizes in HTCS's. Our analysis is based on those reports where the authors used transmission electron microscopy (TEM), HREM or Mössbauer spectroscopy for the determination of the sizes of columnar defects. We will not use the data obtained by the study of the temperature dependence of the resistance because this is a rather indirect method of the determination of track sizes. We also omit those reports when strings of small spheres and elongated defects formed the track. These data are not suitable for our purposes.

The data that we use in the analysis were obtained with various methods by different groups, on different samples in different experimental conditions. Therefore, they cannot be used for the analysis before some selection and corrections are made. It is typical for most of the HREM data that the distribution of track sizes is not given, only the range of variation is reported. It is often omitted what the position of the electron microscopic sample was in the irradiated specimen and what the depth was where S_e was calculated and also what density value was used. When we compare the results of different authors we cannot reduce the effect of these sources of error.

As it was shown by Zhu *et al.*[9] track formation in Y-Ba-Cu-O is rather sensitive to the oxygen content of the samples. Therefore, δ=constant would be desirable for all data. However, if only data for samples with $\delta=0$ were used in the analysis this would significantly reduce their number. Thus, $\delta \leqslant 0.1$ was also allowed that might slightly increase the scatter of points.

The track size is also sensitive to the direction of the irradiation relative to the crystallographic axes.[9] Since most of the results were obtained with irradiation in the *c* direction, therefore other data were rejected when the evolution of track size with S_e was analyzed.

Meftah *et al.*[6] reported that the damage cross section in YIG varies with the ion velocity. Recently, we showed that this effect is related to the variation of the efficiency of energy deposition *g* for ions within the range of 2.2 MeV/nucleon $<E<$7.6 MeV/nucleon.[7] Previously, the velocity effect has not been studied in HTCS's. In the present analysis we did not observe any systematic difference between tracks induced by low and high velocity ions. Thus, it was not necessary to reduce further the number of data points for this reason. As a result of the above considerations, track data in Y-Ba-Cu-O reported in Refs. 10–19 are used in the following analysis.

According to our model, $T_o = T_m - T_{\rm ir}$ is an important parameter in a track formation experiment. The value of the threshold electronic stopping power S_{et} is proportional to T_o. Therefore, the R_e^2-S_e curves are shifted along the S_e axis for samples with different irradiation temperatures. This directly follows from Eqs. (1)–(3). Most of the irradiation experiments on Y-Ba-Cu-O were made at 300 and 80 K, so T_o is not identical even for all samples with $\delta=0$. However, this can be simply taken into account by Eq. (3). In general, S_{et} is higher at a lower irradiation temperature, because higher energy is required to reach the melting point. We chose $T=80$ K for the reference temperature, therefore for $T_{\rm ir}>80$ K all S_e values should be increased, correspondingly. The specific heat at $T=80$ K was taken from Ref. 20 and the Dulong-Petit value was used above 300 K. The temperature dependence of *c* was approximated by a linear variation between 80 and 300 K. In Fig. 1 the track data for Y-Ba-Cu-O are plotted after the correction of S_e to $T_{\rm ir}=80$ K. The original data are also shown in the figure. The correction is about 10% that cannot be neglected, even if the error of S_e data is usually higher than this value.

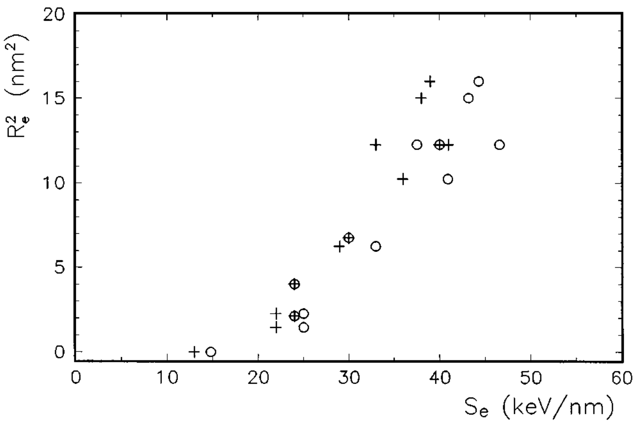

FIG. 1. Track sizes in Y-Ba-Cu-O versus the original (+) and the corrected to $T_{\rm ir}=80$ K (o) S_e values.

In Fig. 2 the R_e^2 data versus the corrected S_e values are plotted. In spite of the fact that most of the data were obtained by different groups, the scatter is not very large. The line in the figure was obtained by a least-squares fit of Eq. (1). The threshold value S_{et} can be determined with rela-

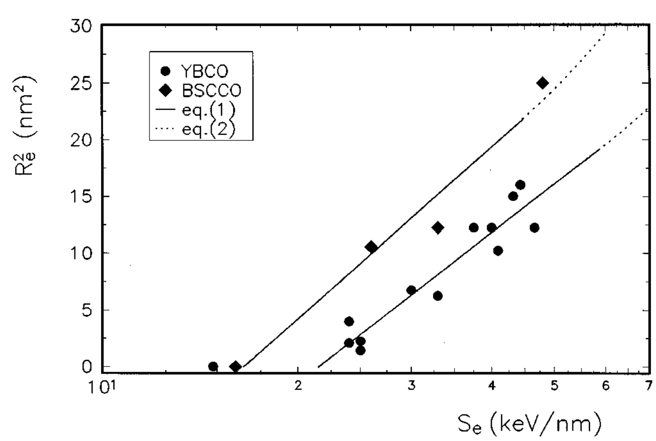

FIG. 2. Variation of the size of columnar defects in Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O:2212 crystals irradiated by swift heavy ions. The curves were calculated by the least-squares method.

tively low uncertainty and $S_{et}=21.5$ keV/nm is found at $T=80$ K that corresponds to $S_{et}=19$ keV/nm for irradiations at 300 K. For the other parameter $a(0)=4.4$ nm was obtained that is surprisingly close to the value found for insulating materials.[4]

There are a few results on track sizes in Bi-Sr-Ca-Cu-O:2212 superconductor[21–24] which are also plotted in Fig. 2. All irradiations were made at liquid nitrogen temperature, so temperature correction was not required. For Bi-Sr-Ca-Cu-O $a(0)=4.65$ nm and $S_{et}=16.5$ keV/nm was provided by the analysis in good agreement with the threshold value (16 keV/nm) reported by Leghissa *et al.*[21] There is a slight difference between the slopes of the two lines in Fig. 2. One should remember that the original data have an experimental error of about $\pm 20\%$, and there are only a few data for Bi-Sr-Ca-Cu-O.

IV. DISCUSSION

Under the conditions used in the experiments the nuclear stopping power can be completely neglected compared to S_e. When an energetic ion penetrates into a solid its energy is transferred through the electromagnetic interaction between the highly charged ion and the electrons. In pure metals the electron thermal diffusivity is high. Therefore, it is expected, that the transferred energy is spread very quickly over the electron system and the electron-phonon coupling leads to a broad spatial temperature distribution with only small peak temperature T_p in the phonon system.[25] In insulating materials there are no conduction electrons and a narrow distribution with high peak temperature is formed in the lattice.[26] Therefore, in insulating materials a low S_{et} value and small tracks $[R_o \sim a(0)]$ are expected, whereas in metals large S_{et} values and large tracks are expected, while parameters for semiconducting specimens have intermediate values. The predicted large tracks for metals may be surprising but the ''bottleneck'' for the metallic systems is the high value of S_{et} required to reach the melting point. For equal S_e/S_{et} values larger tracks are expected in metallic materials than in insulators if amorphization is possible in the given metallic sample.

To compare the track sizes appropriately, we use a normalized S_e/S_{et} plot in Fig. 3 where data for Y-Ba-Cu-O, Bi-Sr-Ca-Cu-O:2212, $NiFe_2O_4$ (Ref. 27) and mica[5] are depicted. Surprisingly, when the track sizes in insulators and in Y-Ba-Cu-O or Bi-Sr-Ca-Cu-O are compared we do not see much difference. The data agree within the experimental error. The solid line is not the result of a fit. It is calculated according to Eqs. (1), (2) using $a(0)=4.5$ nm obtained in Ref. 4 for magnetic insulators. Although the least-squares method provides a better fit for Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O, we do not observe a systematic deviation from the line determined from the analysis of magnetic insulators. The slight differences between the lines in Figs. 2,3 are covered by the scatter of experimental data. Thus, the Gaussian widths of the thermal spike $a(0)$ in the insulating $NiFe_2O_4$, mica and in the HTCS's Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O are equal within the experimental error. It was observed previously that track formation in Y-Ba-Cu-O is similar to that in insulating materials.[28] Our analysis proves this quantitatively.

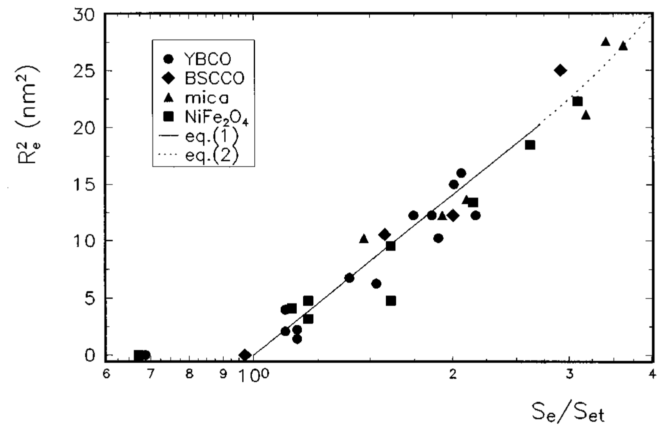

FIG. 3. Normalized plot of track sizes in different materials. The theoretical curve was calculated with $a(0)=4.5$ nm (Ref. 4).

While the $a(0)$ values are in good agreement for Y-Ba-Cu-O, Bi-Sr-Ca-Cu-O and insulating materials, there is a significant difference in the efficiency of energy deposition in the thermal spike g. Its value for Y-Ba-Cu-O can be calculated from Eq. (3). If $\rho=6380$ kg/m^3,[29] $T_o \approx 1070$ K,[30] $T_{ir}=300$ K, $S_{et}=19$ keV/nm as above and the specific heat is calculated analogously to estimating the temperature correction then $g=0.066$ is obtained. A similar calculation leads to $g=0.071$ for Bi-Sr-Ca-Cu-O:2212 ($\rho=6540$ kg/m^3,[15] $T_o \approx 1040$ K,[31] $S_{et}=16.5$ keV/nm, $T_{ir}=80$ K). The results of our analysis show that if the physical properties of the HTCS's are properly taken into account according to Eq. (3), then both thermal spike parameters $a(0)$ and g have very close values in Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O. This agreement confirms the consistency of the results and it provides a further evidence of the formation of a thermal spike along the ion trajectories.

Compared to $g=0.17$ found in magnetic insulators and mica, the $g \approx 0.07$ is a much lower value. The low value of g has important consequences for the production of columnar defects in Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O. The relatively high value of S_{et} is the result of the low efficiency. For $g=0.17$ the S_{et} values would be reduced by about 60% for these HTCS's.

The thermal spike model predicts that $a(0)$ increases in the order of insulator-semiconductor-metal. This prediction is well fulfilled for GeS, the only semiconductor where latent tracks were systematically studied,[32] since $a^2(0) \gg 20$ nm^2 for GeS.[33] However, the value of the parameter g is very close to that in magnetic insulators. In HTCS's the result is the opposite: the $a(0)$ value agrees with that for insulating materials, however, the efficiency g is lower by 60%. The $a(0)$ parameter is related to the average width of the electron energy density distribution during the time interval when the electron energy transfer is the most efficient, therefore a correlation with the electron thermal diffusivity is expected. Because of the low $a(0)$ value the case of Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O apparently contradicts what is expected for a metallic material. Thus, with respect to amorphous track formation these HTCS's exhibit an insulatorlike behavior. A possible explanation is related to the complex nature of HTCS's. In Y-Ba-Cu-O the electron and phonon contributions to the thermal diffusivity in the (ab) planes are nearly equal above T_c, while the electron contribution is negligible compared to the phonon contribution along the c direction.[34] The ratio of the thermal conductivities along the (ab) planes and in the c direction is about 10. Thus, in spite of the metallic conduction in the c direction, as a result of the low electron contribution to the thermal diffusivity the behavior of Y-Ba-Cu-O is similar to that of an insulator for heat transfer in this direction. On the other hand, it is like a poor conductor for the heat transfer along the (ab) planes.

All estimates lead to the conclusion that compared to insulators a much broader thermal spike is expected in materials with high electron thermal diffusivity. The insulatorlike variation of track sizes shown in Fig. 2 indicate that in these HTCS's the electron scattering in the c direction is the most important for track formation. On the other hand, track formation cannot be identical in insulators and HTCS's, because in this case the high electron mobility in the (ab) planes would not have any contribution in the energy transfer, and this is certainly not the case.

A possible solution of this problem can be found if the g values are included in the consideration. The low g values indicate that not all electrons equally participate in the energy transfer to the lattice. It is generally accepted that the energy of the primary electrons is quickly thermalized and the energy of the secondary electrons is about some eV when the electron-phonon scattering becomes efficient. We suppose that the energy distribution of these low-energy electrons is isotropic because after a great number of collisions the electrons lose information about the initial ion velocity. The narrow width of the spike shows that the energy-transfer process leading to amorphous track formation is characteristic to the electron scattering in insulators. Therefore, we compare the g values in HTCS's to $g=0.17$ found in insulators irradiated by high velocity ions.[4] The low g values in HTCS's indicate that in average roughly 1/3 of the electron energy is to be considered in the track formation process for irradiations in the c direction.

We characterize the energy deposition to the lattice by the mean thermal diffusion length $\lambda=(D_e\tau_a)^{1/2}$, where D_e and τ_a are the electron thermal diffusivity and the electron-atom relaxation time, respectively. The λ parameter was used by Toulemonde and co-workers when they evaluated the temperature increase induced by a swift heavy ion in a solid target.[35] For a cylindrical symmetry λ characterizes the size of the cylinder in which the electron energy is deposited to the lattice atoms.

Due to the anisotropic electron scattering in HTCS's, the electron thermal diffusivity is high in the (ab) plane (m-type scattering) compared to that in the c direction (i-type scattering). Consequently, λ is larger in the (ab) plane than in the c direction. Because of the isotropic energy distribution in average 2/3 of the electron energy is transferred to the lattice by m-type scattering. This contribution induces a broad thermal spike with a low peak temperature if irradiation is parallel to the c direction. The process provides a small contribution to the track formation because only a small fraction of the energy transfer is within the track region. The spike temperature may exceed the melting point due to the narrow temperature distribution and high peak temperature coming from the rest of the energy that is transferred to the lattice by i-type scattering. Essentially, a narrow thermal spike is superposed on a broad one. This consideration explains the circular shape of the track, the low efficiency of track formation and the low $a(0)$ value for irradiations in the c direction.

The shape of the track and the efficiency of track formation must be changed when the irradiation is performed in the a direction. As previously, we suppose that 1/3 of the energy of the electron system is deposited close to the trajectory by i-type scattering. Due to the isotropic energy distribution and a high λ value in the b direction, in average 1/3 of the electron energy is taken away from the track region by m-type scattering.

Compared to the previous case, now the beam propagates in a direction where λ is high. However, this does not lead to a strong spreadout of the deposited energy from the track region, because the a direction is perpendicular to the plane of the track. As a result, up to 2/3 of the energy of the electron system may be effective in the process of track formation if HTCS's are again compared to insulators. An elongated track is expected along the b direction for such irradiation since λ is smaller in the c direction than in the (ab) plane. The metallic conduction in the (ab) plane is not crucial for the above consideration. However, it is important that the $a(0)$ values must be very different along the c and other axes. Thus, a qualitatively similar picture is expected for a sample which is semiconducting along the (ab) planes.

Circular tracks and $g\approx0.07$ were found in HTCS's irradiated in the c direction. We predicted a higher efficiency for a beam parallel to the a/b direction and an elongated shape of the tracks in the b/a direction. Both conclusions were experimentally confirmed by the measurements of Zhu *et al.*[9] They irradiated monocrystalline Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O:2223 samples by energetic ions along the c and a/b directions. After ion irradiation columnar defects were observed by HREM. In agreement with our prediction, the cross sections of the defects were circular and elongated for irradiations in the c and a/b directions, respectively.

Zhu *et al.* also observed that besides the shapes the cross section of the tracks also varied with the beam direction. When irradiated in the a/b and c directions the ratio of the elliptical and circular track cross sections were 1.5 and 2.1 for Y-Ba-Cu-O ($\delta=0.7$) and Bi-Sr-Ca-Cu-O:2223, respectively. These results were demonstrated by HREM pictures, which are in qualitative agreement with our prediction regarding the variation of the track cross section. However, the reported data are not suitable for a quantitative comparison.

Analysis of the experimental results showed that the thermal spike model correctly describes the variation of track sizes with S_e in HTCS's. The proposed model of track formation offers a reasonable explanation of the insulatorlike behavior of HTCS's and predicts an anisotropy of the efficiency in agreement with the experimental observations, as well.

By lowering the oxygen content of Y-Ba-Cu-O the superconducting behavior disappears and a semiconducting phase is produced. Zhu *et al.*[9] reported larger tracks in samples with high δ values, i.e., larger tracks were formed in a semiconducting sample than in a metallic one in identical experimental conditions. S_{et} was clearly lower for samples with $\delta=0.7$ than those with $\delta=0$. Track sizes were measured for the semiconducting samples only for two S_e values and two points are insufficient to perform a thermal spike analysis. Moreover, the track sizes given by Zhu *et al.* were the mean values for different beam directions. Thus, track data with different efficiency parameters were mixed. This is the reason why even a rough estimate of the spike parameters cannot be deduced.

The decrease of S_{et} in the semiconducting phase compared to that in the metallic one may come from an increase of g. In different insulators $g=0.17$ for high velocity ion beams. A similar value was obtained for semiconducting GeS, as well.[33] For Y-Ba-Cu-O with $\delta \leqslant 0.1$ $g=0.066$ was found and we expect $g>0.066$ for semiconducting Y-Ba-Cu-O samples with $\delta=0.7$. The higher g value means that the deposition of the electron energy along the ion trajectory is more confined compared to samples with $\delta \leqslant 0.1$. The anisotropy of g is preserved at a higher efficiency, as well, only its magnitude is reduced.

It has been found that the discontinuous amorphous phase is formed as strings of spherical and elongated defects when insulators and HTCS's are irradiated slightly above the threshold electronic stopping value. It was also established that these defects are less effective for flux-line pinning in Y-Ba-Cu-O. A strong pinning can be provided only by long columnar defects.[2] It is of practical interest to estimate the lowest S_{eo} value to produce such defects. By applying Eq. (1) we easily find that tracks with $R \geqslant R_{eo}$ can be formed by ion irradiation with $S_e \geqslant S_{eo} = S_{et}\exp\{[R_{eo}/a(0)]^2\}$ where R_{eo} is a critical radius. We found close similarities between track formation in insulators and HTCS's, therefore it is reasonable to accept for R_{eo} the value obtained by Toulemonde *et al.* for YIG.[1] They reported that discontinuous but overlapping cylindrical defects are formed along the tracks when the track radii are in the range 1.8 nm$<R_e<$3.1 nm and long amorphous cylinders are formed when $R_e>$3.1 nm. According to Ref. 2 a strong pinning effect is induced already when overlapping cylindrical defects are present in the sample along the tracks. Therefore, we use in our calculation $R_{eo}=1.8$ nm leading to $S_{eo}=25.4$ keV/nm for Y-Ba-Cu-O ($\delta \leqslant 0.1$) when the beam is in the c direction and the irradiation is made at 80 K. For $T_{ir}=300$ K continuous columnar defects are expected in Y-Ba-Cu-O for $S_e>31$ keV/nm. This is in agreement with the results of Wheeler *et al.*[36] who observed discontinuous defects at $S_e=28$ keV/nm. They also found that columnar defects become continuous between 28 and 35 keV/nm.

In Bi-Sr-Ca-Cu-O-2212 the magnetic microstructure does not consist of flux lines, as in Y-Ba-Cu-O, but of two-dimensional pancake vortices. As a result, different depinning mechanisms are operative in the two systems. So far, the significance of the morphology of the radiation-induced defects for vortex pinning has not been cleared up in HTCS's with pancake vortices.

As it has been discussed, at a given S_e value the size of the columnar defects can be increased (i) by irradiation in a/b direction, (ii) by reducing the oxygen concentration (iii) by increasing the irradiation temperature. As a result of the application of (i)–(iii), S_{et} can be significantly reduced.

V. CONCLUSIONS

Track formation in irradiated HTCS's (Y-Ba-Cu-O, Bi-Sr-Ca-Cu-O) and in insulating materials (YIG, mica, ferrites) have close similarities. The evolution of track sizes with S_e is correctly predicted by the thermal spike model. The analysis showed that the Gaussian width of the thermal spike $a(0)$ is equal within the experimental error in both groups of materials. Compared to insulators, the efficiency of energy deposition g is lower in HTCS's because the energy of the excited electrons is transferred far from the track region in the a/b direction. The track shape and the efficiency of energy deposition varies for different beam directions. The model is suitable to predict the size of columnar defects induced by ion irradiation.

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