High quality $YBa_2Cu_3O_7$ Josephson junctions made by direct electron beam writing

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High-T_c Josephson junctions have been fabricated by direct electron beam writing over $YBa_2Cu_3O_7$ thin-film microbridges, using scanning transmission electron microscope (STEM) with an accelerating voltage of 80–120 kV. Annealing at 330–380 K increases T_c and I_c of the junctions and makes them more stable. In the operating range of a few degrees below T_c, the junctions show 100% magnetic field modulation of the critical current, microwave-induced Shapiro steps oscillating according to the resistively shunted junction (RSJ) model, and RSJ current-voltage characteristics with I_cR_n product up to 0.5–0.6 mV at 75 K and 0.3 mV at 77 K.

In spite of impressive progress in the fabrication of high-temperature superconducting (HTS) Josephson junctions of various types, problems of nonuniformity, low reproducibility, reduced values of I_cR_n, and especially technological complexity have not been completely overcome. In this letter, we report on the properties of planar Josephson junctions fabricated by the technologically attractive method of direct electron beam writing. The idea of using a focused electron beam for producing weak link structures in HTS films was proposed some time ago,[1,2] and preliminary results have recently been reported.[3,4] This method makes use of the natural properties of HTS materials, such as their high normal state resistivity and sensitivity to disordering, while avoiding complex and less reproducible technological steps, such as creation of grain boundaries, or HTS-normal metal interfaces.

The 25 nm $c_\perp$-oriented $YBa_2Cu_3O_7$ film, prepared on $LaAlO_3$ substrate by the BaF_2 process,[5] was patterned into several 2–4 μm bridges using standard photolithography. The 120 keV electron beam of the Philips CM-12 electron microscope was used to produce a narrow damaged region across the microbridge (see the inset in Fig. 1). Computer controlled writing was done by stopping the beam in a large number of equidistant points, about 2 nm center to center, for a certain dwell time (0.5–4 s). The beam current was approximately 0.6 nA, and the beam on the film surface consisted of a 1 nm bright central spot surrounded by a halo about 10 nm in diameter.

The bridges before writing had T_{c0} of 90.5 K, critical current density $j_c=2\times10^6$ A/cm^2 at 77 K and $j_c\geqslant2\times10^7$ A/cm^2 at 4.2 K, and showed no signs of intrinsic weak links. After e-beam writing the $R(T)$ curve develops a "foot", and the junction becomes superconducting at a lower temperature $T_c<T_{c0}$ (see Fig. 1). Both T_c and foot resistance depend on the writing conditions, so that T_c decreases (down to zero) and the foot resistance increases with irradiation dose. The width of the damaged region was estimated to be about 20 nm,[4] which is consistent with the beam size.

After e-beam writing the microbridges display all the properties of Josephson junctions, i.e., nearly classical magnetic field dependence of the critical current, microwave-induced Shapiro steps, oscillating with microwave power according to the RSJ model, and thermal noise rounded RSJ I-V curves. As-prepared junctions are perfectly stable when stored at 77 K, but change their parameters over several weeks at room temperature. To make the junctions more stable (in terms of $\delta T_c/T_c$ and $\delta I_c/I_c$) over a period of time and also to increase T_c and I_c, we annealed the junctions at room temperature or slightly higher. Figure 2 shows $I_c(T)$ dependencies for the undamaged bridge and for the junctions before and after annealing at 300 and 348 K. The relative change of T_c and I_c decreases with increasing annealing time. Short annealing at 348 K gives the same result as a long one at 300 K (Fig.

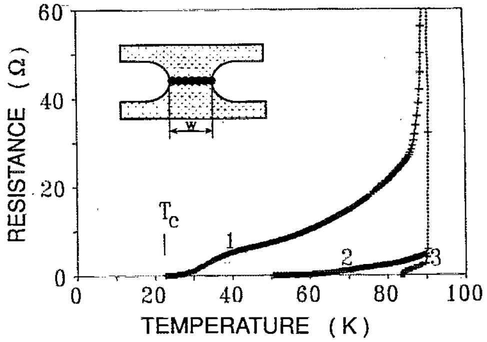

FIG. 1. $R(T)$ dependencies for e-beam treated microbridges. (1)-sample No. 1 ($w=2$ μm, 1.5 s dwell time, 1.5 nm stop-point separation); (2)-sample No. 2 ($w=4$ μm, 2 s dwell time, 2.4 nm stop-point separation); (3)-sample No. 2 annealed at 300 K for 3 months. Inset shows schematically the e-beam treated bridge.

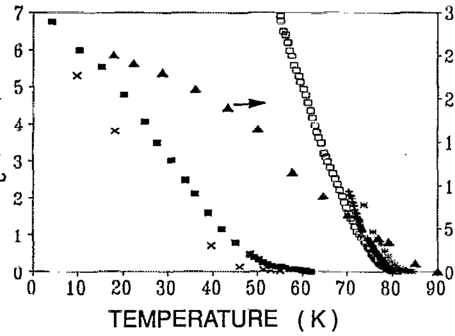

FIG. 2. Temperature dependence of the critical current (0.5 μV criterion). (Δ)-initial 4 μm bridge; ($\times$) sample No. 2 right after writing; (+) No. 2 annealed at 300 K for 2 months; (*) No. 2 annealed at 300 K for 3 months; (■)-sample No. 3 ($w=3$ μm, 1 s dwell time, 1.8 nm stop-point separation) right after writing; (□) No. 3 annealed at 348 K for 30 min.

2). The rate of time variation of the junction parameters was found to be proportional to $\exp(-\sqrt{t/\tau})/\sqrt{t/\tau}$, where the relaxation time is $\tau=\tau_0 \exp(U/kT)$ with $\tau_0 \cong 4 \times 10^{-12}$ s, and the activation energy $U \cong 1.1$ eV. For the annealed junction with $T_c=83$ K, these numbers give $\delta T_c/T_c \approx 4 \times 10^{-5}$ ($\delta I_c/I_c$ of the same order) in a year, if stored at 273 K. More detailed results of annealing experiments will be published separately. We note that the Josephson measurements provide unprecedented sensitivity for studying oxygen ordering and diffusion in HTS.

Figure 3 shows the typical *I-V* curve, magnetic field dependence of I_c, and the modulation of I_c in a 9.3 GHz microwave field for the room-temperature annealed junction. The *I-V* curve can be fitted well by the RSJ model (see Fig. 3). The rounding near $V=0$ corresponds to the noise parameter $\gamma \approx 0.1$, in good agreement with that expected for thermal noise, where $\gamma = 2\pi k_B T / I_{c0} \Phi_0$, and I_{c0} is a noise-free critical current. At temperatures close to T_c, magnetic field modulation is always 100%, implying good uniformity of the damaged region. The characteristic voltage of a junction V_c (that is $I_{c0}R_n$ for the noise-free RSJ shape of the *I-V* curve) can be calculated from the ratio $I_{\rm rf}^{(2)}/I_{\rm rf}^{(1)}$ using results of the RSJ model, where $I_{\rm rf}^{(1)}$ and $I_{\rm rf}^{(2)}$ correspond to the first and the second zero in the $I_c(I_{\rm rf})$ dependence.[6,7] Obtained V_c values agree well with $I_{c0}R_n$ values found from the RSJ fit of the *I-V* curves, and serve as a fine measure for the characteristic frequency of the junction. We note that the widely used I_cR_n criterion fails in the presence of noise or excess current.

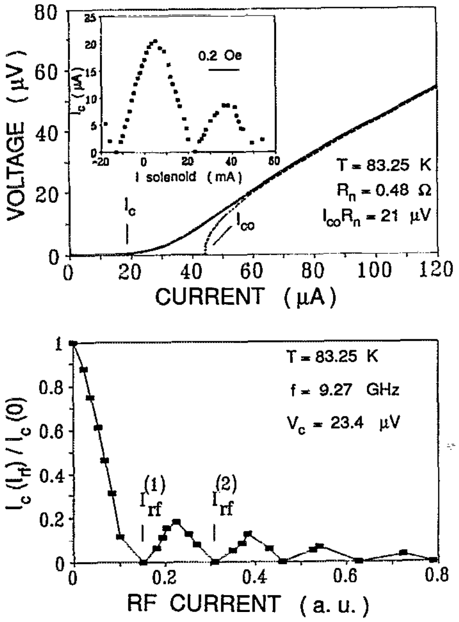

FIG. 3. *I-V* curve, magnetic field modulation and microwave current dependence of the critical current for the sample No. 2 after 3-month annealing. Dashed line is the noise-free RSJ model fit.

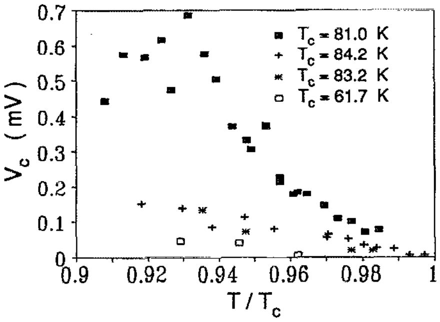

FIG. 4. Temperature dependence of the junction's characteristic voltage, V_c. (□) No. 3 right after *e*-beam writing; (■)- No. 3 after annealing at 348 K for 30 min; +-No. 2 after 2-month annealing at 300 K, (*)-after 3 month annealing at 300 K.

Near T_c, the critical current of *e*-beam treated bridges varies as $I_c = \alpha T_c (T_c - T)^2$ (see Fig. 2), which is characteristic for *SNS* junctions.[8] The normal resistance R_n is in the range of 0.5–1.5 Ω. At lower temperatures (usually a few degrees below T_c) $I_c(T)$ curves deviate from the above relation, while *I-V* curves gradually change shape from the RSJ to the flux-creep shape similar to that found in the original nonirradiated bridge. Nonetheless, Shapiro steps are always observed down to 4.2 K. The change in *I-V* curves at low temperatures can be caused by the transition from short to long (in comparison with the Josephson penetration depth) junction, or from *SNS* to *SS'S*. We leave this issue for a more detailed publication.

Figure 4 shows the temperature dependence of V_c (from the microwave measurements) before and after annealing. The change of the shape of *I-V* curves, mentioned above, causes an uncertainty in determination of the junction's parameters, which results in the scattering of points in Fig. 4 at low temperatures. Note that V_c can be maximized by choosing an optimum annealing time because T_c and I_c increase, and R_n decreases with annealing. However, even without this optimization, V_c values obtained as good as any of the published I_cR_n values at 77–83 K for other junction technologies.

In summary, we have found a simple and technologically attractive method of reproducible fabrication of $YBa_2Cu_3O_7$ planar Josephson junctions operating in the range of 77–85 K, which consists of direct electron beam writing across thin-film microbridges and subsequent annealing at 330–380 K. The junctions are most likely of *SNS* (*SS'S* at low temperatures) type with $V_c \approx 0.5$ mV at $T/T_c = 0.93$ and 0.3 mV at 77 K. For long term stability (of the order of one year, or over the duration of a grant), the junctions must be stored at $T \leqslant 273$ K. Typical electron beam writing time in our microscope is 5–10 min per μm which allows fabrication of simple multijunction circuits. This time can be reduced by orders of magnitude in a microscope with significantly higher beam current. The method, when applied to high quality uniform films, appears to have a potential for good uniformity and reproducibility from run to run. Detailed study of these parameters is in progress. The method also opens up the possibility of direct *e*-beam patterning on a very fine scale, e.g., for creation of SQUIDs with submicron size loops, or Josephson arrays with a submicron distance between junctions. Multilayered structures can also be envisioned with the junctions produced at the last stage of a technological process.

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