

the tossing of a fair coin, there is no algorithm which can shorten the sequence' because the sequence might be all heads, for example. Bernoulli's *Ars conjectandi* contained no discussion of actual sequences, and his name should surely be reserved for the generating process.

But, as Yockey goes on to say, "It is impossible to prove that any given sequence was generated by a random process"; in other words, it is impossible to prove that a process is random by examining its output sequence. Rather, a process is called random if its structure ensures that its output is unpredictable.

Statisticians are familiar with the separate uses. A good experimental design will need to be based on the first kind of randomness lest it display systematic bias, but a randomized trial will need the second kind so that the participants cannot guess what is coming next.

Whether randomness can be measured is a difficult problem (*Nature* **344**, 705; 1990). One cannot judge the absence of pattern without specifying which pattern, and what is a pattern to you may not be a pattern to me.

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## Superconductor defect structure

SIR—One of the main obstacles to the practical use of high-temperature superconductors is that the amounts of current that they can carry and still remain superconducting (that is, their critical current densities) are relatively low. A processing method that has significantly increased the intragranular critical current density involves the solid-state phase transformation of single-phase  $YBa_2Cu_3O_{7-x}$  ('124' or '248') into  $YBa_2Cu_3O_7$  ('123') with the concomitant formation of a heavily defective microstructure. Here we report a defect structure of apparent composition  $YBa_2Cu_3O_7$  ('125') in solid-state-transformed sintered bulk samples of 124.

The decomposition of 124 precursor was carried out by heat treating it at 930 °C for 1.5 min followed by a low temperature oxygenation process of rapidly cooling (10 s) to about 750 °C with further cooling to 380 °C at 20 °C h<sup>-1</sup> in flowing oxygen. Further details of sample preparation are given elsewhere<sup>1</sup>. The defect structure was examined by high-resolution transmission electron microscopy (HREM) using the Berkeley atomic-resolution microscope. Thin foils were prepared by mechanically grinding a slice of the superconductor to about 50–80 μm, followed by argon-ion milling

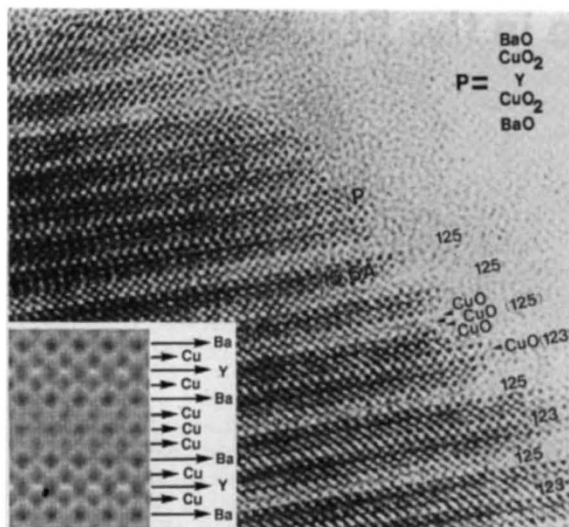


FIG. 1 Atomic-resolution image of the phase-decomposed 124 sample illustrating the 123 structure alternating with the new 125 defect structure. Layers of 125 two or three unit cells thick are seen in the upper and middle part of the micrograph. The simulated intensity distribution for the cation positions (inset) of 125 for a foil thickness of 12 Å and an objective-lens defocus of -500 Å, confirms the interpretation of the contrast in the micrograph.

to electron transparency at 6 kV and 77 K. Images were obtained under conditions close to Scherzer defocus and the projections of atomic columns therefore appear as dark dots on a white background. To avoid electron-beam-induced damage, images were typically recorded within 2 min of exposure to the beam.

A micrograph of the defect structure (Fig. 1) shows the decomposition of 124 into 123 and 125. The structures of the three related phases are illustrated in Fig. 2 (a–c). Analysis of the image contrast using detailed image simulations indicates that the basic perovskite building blocks (marked P in Fig. 1) in the 125 structure are similar to those in the 123 structure, except that the 125 structure has three CuO chain layers whereas 123 has one. The 124 structure has two CuO chain layers with an  $a/2[100]$  glide plane between

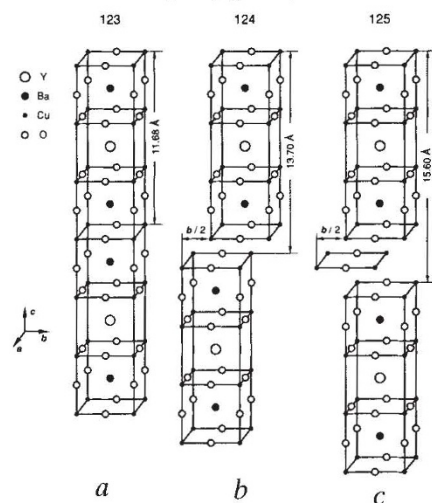


FIG. 2 Schematic illustrations of the structure 123, 124 and 125.

them<sup>2,3</sup>. In the proposed 125 structure (most likely  $YBa_2Cu_3O_9$ ) the three CuO chains are mutually related by a  $1/2[100]$  glide operation and hence, unlike 124, there is no doubling of the unit-cell dimension along the  $c$  direction. The  $c$ -axis lattice parameter is expected to be 15.6 Å, based on the addition of one more CuO chain layer to the 124 structure (the Cu–O distance along the  $c$  axis is about 1.9 Å, as established by neutron diffraction and HREM analysis<sup>2</sup>). In general, single layers of 125 alternate with single layers of 123 but occasionally layers of 125 two to three unit cells thick occur.

The fact that the 123 and 125 layers are formed as alternating layers with very rapid kinetics suggests that the early stage of the

decomposition of 124 may occur by a spinodal (spontaneous decomposition without a nucleation barrier) process. Such a decomposition to 123 requires only short-range diffusion along the  $c$  direction (jumps over atomic-layer distances), as can be seen from Figs 1 and 2. During the later stage of decomposition, however, nucleation and growth reactions seem to take over, involving the removal of copper and oxygen by long-range planar diffusion to form equilibrium, twinned 123 and spherical CuO inclusions.

So far, our attempts to synthesize the 125 phase as a single phase in bulk form by conventional ceramic processing have failed. Perhaps the formation of the 125 phase is easier when the 124 phase spontaneously decomposes into the 123 structure, forcing the formation of the 125 structure. We note that recent three-dimensional Monte Carlo simulations using experimental thermodynamic data predict that a bulk 125 containing material can be synthesized at a very high oxygen pressure in a copper-rich environment (M. Fendorf, personal communication).

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