Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

"Universal relationship between sample dimensions and cooperative properties: Effects of fractal dimension on electronic properties of high-T_C cuprate observed using electron spin resonance"

by

Toshio Naito and Yoshiaki Fukuda

1. Derivation of $V = V(m, n, l)$

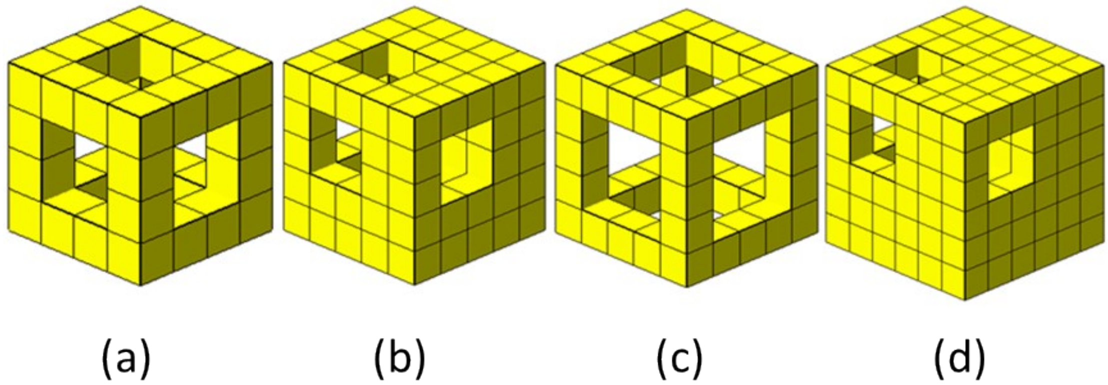

Fig. S1. Fractal models of the first generation; $(m, n, l) = (1, 4, 2)$ **(a)**, $(1, 5, 2)$ **(b)**, $(1, 5, 3)$ **(c)**, and $(1, 6, 2)$ **(d)**.

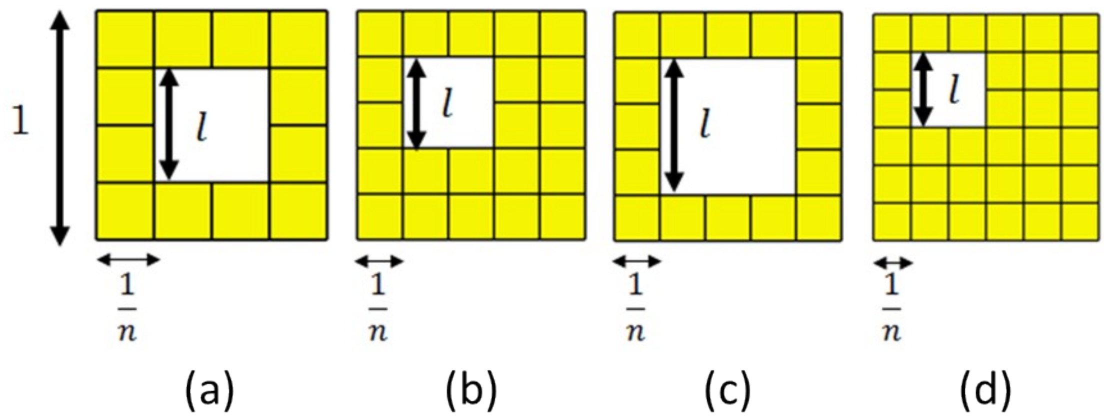

Fig. S2. Side views of the fractal models of the first generation in Fig. S1; $(m, n, l) = (1, 4, 2)$ **(a)**, $(1, 5, 2)$ **(b)**, $(1, 5, 3)$ **(c)**, and $(1, 6, 2)$ **(d)**.

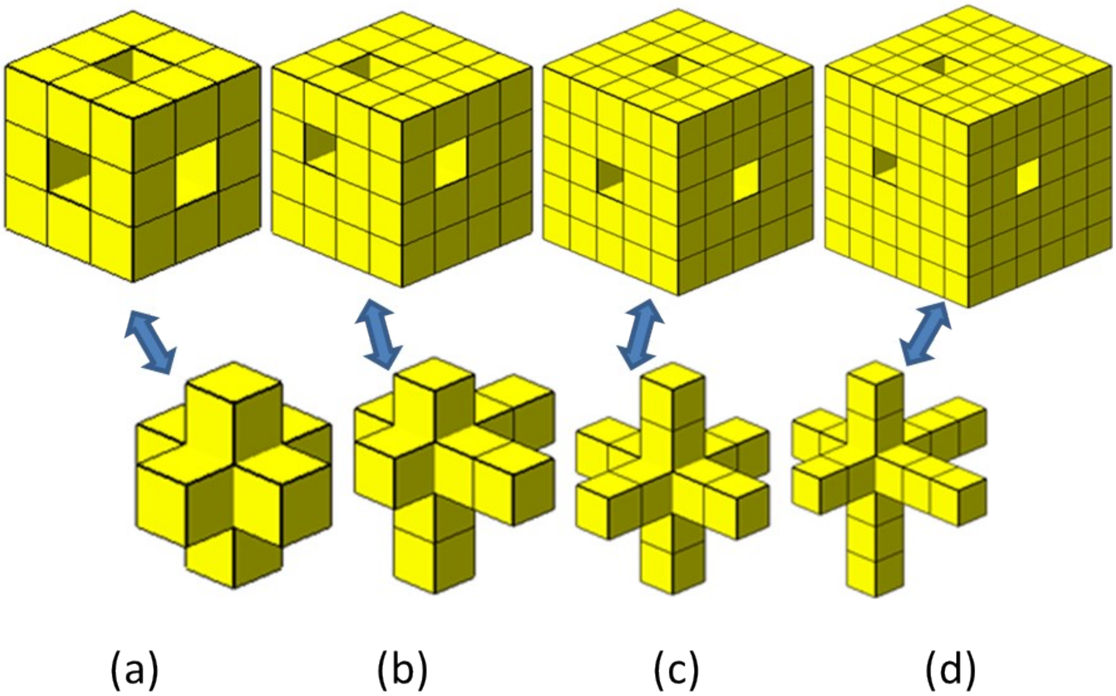

Fig. S3. Removed (lower figures) and residual (upper figures) parts of the fractal models of the first generation; (m, n, l) = (1, 3, 1) **(a)**, (1, 4, 1) **(b)**, (1, 5, 1) **(c)**, and (1, 6, 1) **(d)**.

At first, a cube with a unit length of edge is divided by n^3 cubes of equal volumes, producing small cubes with an edge of n^{-1}. Then we remove some of the small cubes to leave square holes with an edge of l in the orthogonal three directions (Figs. S1 and S2; $1 \leq l \leq n-2$).

The volume to be removed from the original (intact) cube for the first generation fractal models, V_r, is calculated from the volume of the square pillar with considering the duplicated central part (Fig. S3).

$$V_r = \frac{1}{n^3} \times \left(3 \times n \times l^2 - 2l^3\right) = \frac{l^2(3n-2l)}{n^3} \qquad (Eq.S1)$$

Then, the residual volume, *i.e.*, $V(1, n, l)$ is obtained by subtracting V_r from the original volume (=1).

$$V(1,n,l) = 1 - \frac{l^2(3n-2l)}{n^3} = \frac{1}{n^3}\{n^3 - l^2(3n-2l)\} \qquad (Eq.S2)$$

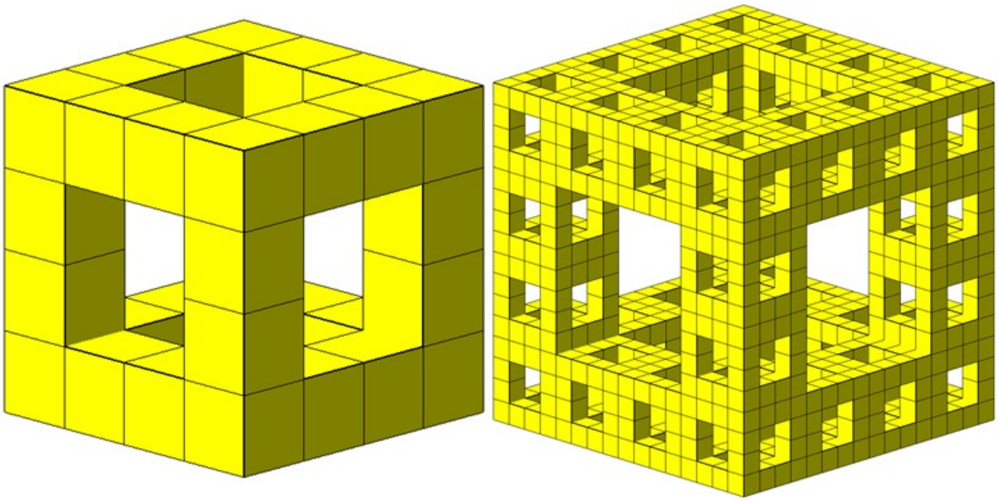

Fig. S4. Relationship between fractal models with different generations; the first generation; $(m, n, l) = (1, 4, 2)$ **(a)** and the second generation; $(m, n, l) = (2, 4, 2)$ **(b)**.

The volume of the second generation $V = V(2, n, l)$ is calculated from that of the first generation $V = V(1, n, l)$ (Fig. S4(a)). Every smallest cube in the first generation is divided into n^3 cubes of equal dimension. Then we remove some cubes to leave holes in the exact same way with what we did in the first generation (Fig. S4(b)). Therefore, the volume $V(2, n, l)$ is described by (Eq. S3).

$$V(2,n,l) = \frac{1}{(n^2)^3}\{n^3 - l^2(3n - 2l)\} \times \{n^3 - l^2(3n - 2l)\} = \frac{1}{(n^2)^3}\{n^3 - l^2(3n - 2l)\}^2$$

(Eq. S3)

If we repeat a similar procedure for the fractal model of the second generation, we obtain the volume of the third generation $V(3, n, l)$.

$$V(3,n,l) = \frac{1}{(n^3)^3}\{n^3 - l^2(3n - 2l)\}^2 \times \{n^3 - l^2(3n - 2l)\} = \frac{1}{(n^3)^3}\{n^3 - l^2(3n - 2l)\}^3$$

(Eq. S4)

Similarly, the volume of the m-th generation $V = V(\mathrm{m}, n, l)$ is described by (Eq. S5).

$$V(m,n,l) = \frac{\{n^3 - l^2(3n - 2l)\}^m}{(n^m)^3} \; (m = 0,1,2,\ldots;\ 1 \le l \le n - 2) \qquad (Eq. S5)$$

2. Derivation of $S = S(m, n, l)$

Firstly, we derive the surface area of the first generation. The side product of a given fractal model (Fig. S2) is

$$\frac{1}{n^2} \times (n^2 - l^2) \qquad (Eq. S6)$$

As every fractal model has six sides, the surface area of all the sides S(sides) of the first generation is

$$S(sides; m = 1) = \frac{6}{n^2} \times (n^2 - l^2) \qquad (Eq. S7)$$

Next we consider the inner surface area S(inner) of the first generation fractal models.

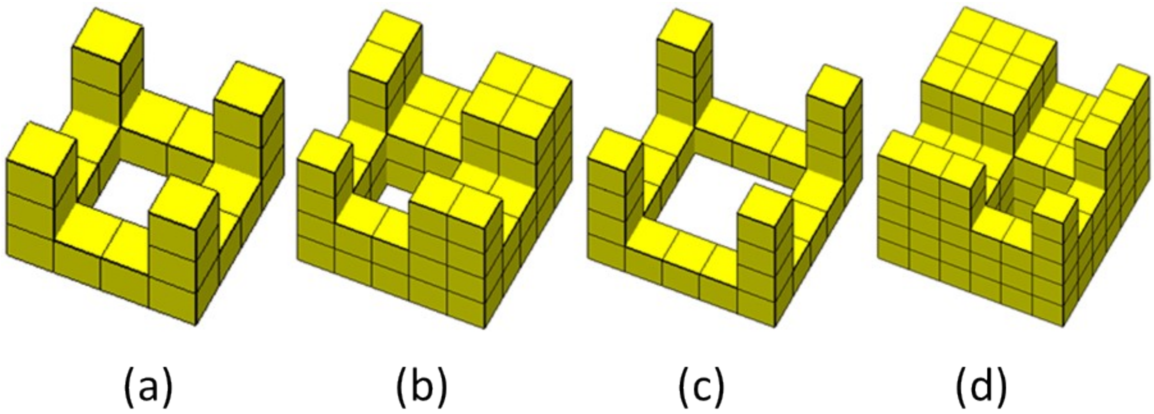

Fig. S5. Inside views of the fractal models of the first generation; (m, n, l) = (1, 4, 2) **(a)**, (1, 5, 2) **(b)**, (1, 5, 3) **(c)**, and (1, 6, 2) **(d)**.

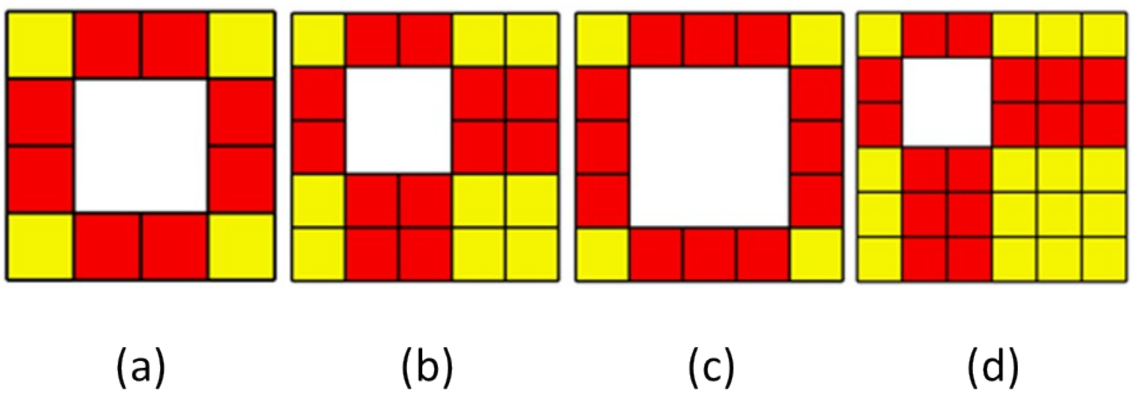

Fig. S6. Projection views of the fractal models of the first generation shown in Fig. S6; (m, n, l) = (1, 4, 2) **(a)**, (1, 5, 2) **(b)**, (1, 5, 3) **(c)**, and (1, 6, 2) **(d)**. The red parts designate the increased surface area by producing the void spaces.

The void space in the first generation fractal model consists of orthogonally interpenetrating square pillars as shown in Fig. S3. The side product of a square pillar is

$$\frac{1}{n^2} \times l \times (n - l) \qquad (Eq. S8)$$

As the void space consists of three pillars with four sides each, S(inner; m = 1) is described as

$$S(inner; m = 1) = 4 \times 3 \times \frac{1}{n^2} \times l \times (n - l) = \frac{12}{n^2} l(n - l) \quad (Eq. S9)$$

Thus the total surface area of the first generation $S(1, n, l)$ is calculated as the sum of S(sides; m = 1) and S(inner; m = 1).

$$S(1, n, l) = \frac{6}{n^2}(n^2 - l^2) + \frac{12}{n^2} l(n - l) \quad (Eq. S10)$$

The second generation surface area $S(2, n, l)$ is calculated similarly to (Eq. S10). The second generation fractal model consists of the smaller cubes with an edge length of n^{-2} (Fig. S7). As a side of the second generation fractal model contains the following number (Eq. S11) of the side of the first generation model,

$$(n^2 - l^2) \times (n^2 - l^2) = (n^2 - l^2)^2 \quad (Eq. S11)$$

the side product of a given fractal model of the second generation is

$$\frac{1}{(n^2)^2}(n^2 - l^2)^2 \quad (Eq. S12)$$

As every fractal model has six sides, the surface area of all the sides S(sides) of the second generation is

$$S(sides; m = 2) = \frac{6}{(n^2)^2}(n^2 - l^2)^2 \qquad (Eq. S12)$$

The number of the smaller holes in the second generation is

$$n^3 - 3n + 2 \qquad (Eq. S8)$$

Every smallest hole has four sides and extends in three directions. Thus the surface area of the smaller holes S(smaller holes) is

$$S(smaller\ holes) = \frac{12}{(n^2)^2} \times l(n - l)\{n^3 - l^2(3n - 2l)\} \quad (Eq. S13)$$

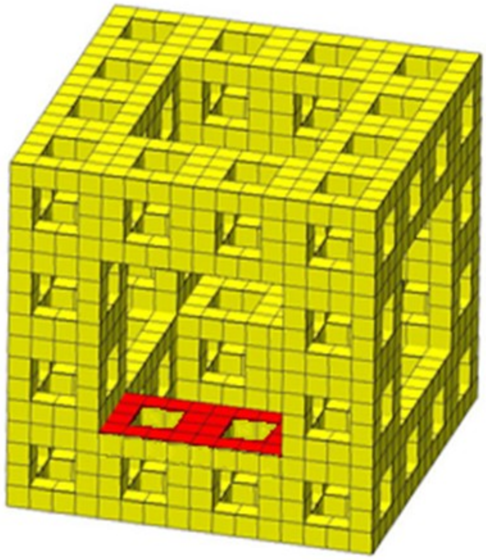

Fig. S7. Fractal model of the second generation; (m, n, l) = (2, 4, 2). The red parts designate a basal plane of the largest void space.

Then we consider the larger holes. The red area in Fig. S7 consists of two (more generally, l) sides of the first generation model. Each side area of the smaller square is $(n^{-2})^{-2}$, and there are (n^2-l^2) of smaller squares in a side of the first generation model. Thus the red area in Fig. S7 is described by (Eq. S14).

$$\frac{1}{(n^2)^2}l(n^2-l^2) \qquad (Eq. S14)$$

Therefore, by similar discussion to derive (Eq. S13), the surface area of the larger holes S(larger holes) is

$$S(larger\ holes) = \frac{12}{(n^2)^2}l(n-l)(n^2-l^2) \qquad (Eq. S15)$$

Thus the surface area of the second generation $S(2, n, l)$ is described by the sum of (Eq. S12), (Eq. S13), and (Eq. S15).

$$S(2,n,l) = \frac{6}{(n^2)^2}(n^2-l^2)^2 + \frac{12}{(n^2)^2}l(n-l)\{n^3-l^2(3n-2l)\} + \frac{12}{(n^2)^2}l(n-l)(n^2-l^2)$$

(Eq. S16)

Similarly, for the m-th generation,

$$S(m,n,l) = 6\frac{(n^2-l^2)^m}{(n^m)^2} + 12\frac{l(n-l)}{(n^m)^2}\sum_{k=1}^{m}(n^2-l^2)^{k-1}\{n^3-l^2(3n-2l)\}^{m-k}$$

$$(m = 0,1,2,...;\ 1 \le l \le n-2) \qquad (Eq. S17)$$

3. The fitting analysis of critical current density performed in our previous work[S1]

The trial function for the observed D_f-dependence of $J_C(H)$ is

$$J_C(H) = J_{C,0} + A_1 exp\left(-\frac{H}{\tau_1}\right) + A_2 exp\left(-\frac{H}{\tau_2}\right) \qquad (Eq. S18)$$

where H, $J_{C,0}$, A_i, and τ_i (i = 1,2) indicate the magnetic field strength [G], offset values of J_C [A cm^{-2}], amplitudes [A cm^{-2}], and relaxation constant [G^{-1}], respectively.

Among the parameters of $J_{C,0}$, A_i, and τ_i, only τ_i (i = 1,2) has a physical meaning intrinsic to the sample and independent of the experimental conditions. The value of $J_{C,0}$ originates from residual magnetization and depends on the amount and distribution of defects in samples and the experimental conditions such as magnetic fields. The absolute values of A_i depend on the system sizes (the volumes of the AKD and YBCO parts). The relative values of A_i, i.e., A_1/A_2 or A_2/A_1, have a physical meaning intrinsic to the fractal dimension D_f. The relative values of A_i describe the ratio of the number of the flux lines penetrating the AKD parts to those penetrating the YBCO parts. Because AKD (a diamagnetic insulator) and YBCO should have clearly different strengths of pinning magnetic flux lines and thus clearly different values of τ_i, the curve-fitting analysis above can distinguish one from the other. Estimated errors in τ_i^{-1}, A_i, and D are approximately ±2.5 %, ±2.5 %, and ±0.05, respectively.

Reference

S1) T. Naito, H. Yamamoto, K. Konishi, K. Kubo, T. Nakamura and H. Mayama, Adv. Mater. Sci., **1**, 15 (2016).