Supplementary material

S1. Approximate distribution function for the case of initially same-sized loops

It is assumed next that in annealing every survived loop has an effective volume V around it. Every loop initially present in the volume V was reacted with the survived one. Prior to annealing, there was a certain random number A of positive and B of negative loops in this volume V with average values, respectively, λ_+ and λ_-:

$$\lambda_{+/-} = Vn_{0,+/-}. \tag{S1.1}$$

Here $n_{0,+/-}$ – initial density of positive and negative loops, respectively. Distributions for A and B (X):

$$P_{\pm}(X) = \lambda_{\pm}{}^{X}\left(X!\right)^{-1}\exp(-\lambda_{\pm}). \tag{S1.2}$$

$$P_{\pm}(1) = \exp(-\lambda_{\pm}). \tag{S1.3}$$

The joint probability that the survived loop absorbed A positive loops and B negative loops:

$$P(A,B) = P_{+}(A)P_{-}(B). \tag{S1.4}$$

If initially all loops had the same radius R_0, the square of the resulting loop radius R^2 is:

$$R^2 = R_0{}^2\left|K\right| = R_0{}^2\left|\left(A-B\right)\right|. \tag{S1.5}$$

And the sign of the loop is the sign of $K = A - B$. Negative area πR^2 is treated as a positive area of negative loop. The probability for the loop to have a radius $R_0{}^2K$ is $P(K)$:

$$P\left(K\right) = \sum_{A,B:(A-B)=K} P(A,B). \tag{S1.6}$$

The calculation of $P(K)$ is easily done numerically. Also, with an accuracy of up to ≈5 % it can be replaced by an integral as it will be done next.

The case $A = B$ results in a fictive loop with zero radius. Therefore, the value V is not just mean volume per loop, but rather an effective recombination volume with the number of survived (i. e., observed) loops n obeying the equation:

$$\left(n + n_{\text{empty}}\right)V = 1, \tag{S1.7}$$

where n_{empty} are the annihilated ones:

$$n_{\text{empty}} = \left(n + n_{\text{empty}}\right)P(K = 0, Vn_0), \tag{S1.8}$$

what results in

$$V = n^{-1}\left(1 - P(K = 0, Vn_0)\right). \tag{S1.9}$$

That is, V is to be found implicitly from this nonlinear equation using the function $P(K = 0, Vn_0)$. The latter has a simple form according to Eq. (S1.6):

$$P(K = 0, Vn_0) = \exp(-\lambda_+ - \lambda_-)\sum_{A=1,\infty}\left(\lambda_+\lambda_-\right)^{A}\left(A!\right)^{-2}. \tag{S1.10}$$

Equations (S1.9) and (S1.10) are solved iteratively for V at a given time t using in each step updated values $\lambda_{+/-} = Vn_{+/-}$ in Eq. (S1.1).

One may use numerical calculation for Eq. (S1.6) with randomized A and B, or proceed with analytical integration:

$$\frac{dP(K)/d(K)}{\exp(-\left(\lambda_+ + \lambda_-\right))} = \int_{B=0}^{\infty}\frac{\lambda_+{}^{(B+K)}}{\left(B+K\right)!}\frac{\lambda_-{}^{B}}{B!}dB = \frac{1}{2\pi}\int_{B=0}^{\infty}\frac{\exp\left\{U(B,K)\right\}}{\sqrt{B\left(B+K\right)}}dB, \tag{S1.11}$$

where

$$U(B,K) = \left(B+K\right)\log\left(\lambda_+\right) + B\log\left(\lambda_-\right) - B\log(B/e) - (B+K)\log\left(\left(B+K\right)/e\right). \tag{S1.12}$$

If $K < 0$ (negative loops) then the integration limit is from $-K$ but not from 0, because $A = K + B$ is an any non-negative number.

https://doi.org/10.48612/letters/2025-4-297-303

An integral of this type is approximately calculated by the saddle-point method, done by expanding the function $f(B)$ in a series near its maximum (at B^* such that $df(B^*)/dB = 0$) to the second order in $(B - B^*)$. The main contribution to the integral occurs close to B^*, so for the remaining functions, their values are taken at the point B^*, since they are much slower functions of B. Integration leads to the "additional error function" erfc (x):

$$\int_{B_1}^{\infty} \exp(f) g dB = g(B^*)\exp(f(B^*))\left(-f(B^*)''/2\right)^{-1/2}\frac{\sqrt{\pi}}{2}\mathrm{erfc}\left(\left(B_1 - B^*\right)\sqrt{-f''/2}\right). \quad \text{(S1.13)}$$

Respectively:

$$f' = 0 \rightarrow B^* = \frac{1}{2}\left(-K + \sqrt{K^2 + 4\lambda_+\lambda_-}\right). \quad \text{(S1.14)}$$

$$f''(B^*) = -\frac{1}{B^* + K} - \frac{1}{B^*} = -\frac{\sqrt{K^2 + 4\lambda_+\lambda_-}}{\lambda_+\lambda_-}. \quad \text{(S1.15)}$$

$$f(B^*) = \frac{1}{2}K\log\left(\lambda_+/\lambda_-\right) + \sqrt{K^2 + 4\lambda_+\lambda_-} + \frac{1}{2}K\log\left(\frac{\left(-K + \sqrt{K^2 + 4\lambda_+\lambda_-}\right)}{\left(K + \sqrt{K^2 + 4\lambda_+\lambda_-}\right)}\right). \quad \text{(S1.16)}$$

Applied in Eq. (S1.2):

$$dP/dK(K > 0) = \exp(-\lambda_+ - \lambda_-)\exp\left\{\sqrt{K^2 + 4\lambda_+\lambda_-}\right\}\left\{\frac{1}{2\sqrt{2\pi}}\right\}\left(K^2 + 4\lambda_+\lambda_-\right)^{-1/4} \times$$

$$\times\left(\frac{\lambda_+}{\lambda_-}\frac{\left(-K + \sqrt{K^2 + 4\lambda_+\lambda_-}\right)}{\left(K + \sqrt{K^2 + 4\lambda_+\lambda_-}\right)}\right)^{\frac{1}{2}K}\mathrm{erfc}\left(-\frac{1}{2}\left(-K + \sqrt{K^2 + 4\lambda_+\lambda_-}\right)\left(\frac{\sqrt{K^2 + 4\lambda_+\lambda_-}}{2\lambda_+\lambda_-}\right)^{1/2}\right). \quad \text{(S1.17)}$$

For $K < 0$ (keeping in mind that here negative K corresponds to a negative-signed loop) the only change in the formula is

$$dP/dK(K < 0) = \ldots \times \mathrm{erfc}\left(-\frac{1}{2}\left(K + \sqrt{K^2 + 4\lambda_+\lambda_-}\right)\ldots\right)\ldots \quad \text{(S1.18)}$$

It is easy to verify that $\lambda_- = \lambda_+$ results in symmetrical SDF in K.

S2. Size distribution function accounting for statistics of recombination volumes

Comparison SDF based on only mean recombination volume with the result of the numerical experiment shows that it doesn't describe the "tail" of the size distribution. It seems that the statistics of recombination volumes is an essential feature for understanding large loop formation. This results in averaging function $P(K, Vn_0)$ under the natural assumption that the recombination volumes Vn_0 are distributed in the same way as the volumes of the empty sphere around the loop, i. e., the previously obtained RCF $\rho'(r')$. Effective volume V is then replaced with $(4/3)\pi r'^3 \cdot V\gamma$ and further convoluted with the function $\rho'(r')$ from Eq. (23). The factor γ comes from the normalization condition: the average volume per loop $4/3\pi r'^3$, must be equal to Vn, i. e.:

$$\gamma^{-1} = \int_{r'=0}^{\infty}\frac{4}{3}\pi r'^3\rho'\left(r'\right)dr' = 1/1.5. \quad \text{(S2.1)}$$

The resulting function is an integral over recombination volume's distribution:

$$P'(K,V) = \int_{r'=0}^{\infty} P\left\{K, \frac{4}{3}\pi r'^3 \cdot VN_0 \cdot \gamma\right\}\rho'\left(r'\right)dr'. \quad \text{(S2.2)}$$

If the initial sizes of the positive and negative loops differ (respectively, R_A and R_B), the procedure remains the same, but the result is more conveniently formulated with a continuous distribution:

$$P\left(\mathrm{Sign}\cdot R^2\right)\Delta R^2 = \sum_{A,B:0<(AR_A{}^2 - BR_B{}^2) - R^2 < \Delta R^2} P(A,B)\left(AR_A{}^2 - BR_B{}^2\right), \quad \text{(S2.3)}$$

where "Sign" is a sign of the loop. This means that sum is taken with any non-negative number for A and B except $A = B = 0$.

An even more realistic situation to be discussed is when the initial width of SDF (size distribution) cannot be ignored. Let the initial distributions $P(s+)$ and $P(s-)$ of the squared radius of positive and negative loops have the mathematical expectation and variance:

$$\mu = \left\langle R^2 \right\rangle; \qquad \text{var} = \left\langle \left(R^2 - \left\langle R^2 \right\rangle \right)^2 \right\rangle. \tag{S2.4}$$

The cases $A=1$, $B=0$ and $A=0$, $B=1$ correspond to the probability that there were no other loops in the effective recombination volume around the loop at the initial moment of time ("+" for positive and "–" for negative x):

$$P_0(\text{Sign}\cdot R^2) = \exp(-Vn_0)\frac{(1\pm\Delta)}{2}\text{norm}(\text{Sign}\cdot R^2, \mu_\pm, \sigma_\pm). \tag{S2.4}$$

where "norm" is the normal distribution. This P_0 term is not convoluted with V (i. e., for Eq. (S2.5) the average value V is used). The first reason is a rapid decrease in P_0 with an increase in $Vn_0 > 1$. The second reason is the distribution $\rho'(r')$ (Eq. (23)) itself is established only at $Vn_0 > 1$. For the remaining sum with $A + B > 1$, each term of the sum in Eq. (S2.3) will be represented as a sum of $S_+ + S_-$, where:

$$S_\pm = \pm X s_\pm, \tag{S2.6}$$

$X = $ A for "+" and $X = $ B for "–" sign. According to the Central Limit Theorem in statistical analysis [10] , for the limit of large a and b, the distribution function of the sum (S2.6) of identically distributed variables tends to a normal distribution with the following parameters:

$$S_\pm \to \text{norm}\left(\pm X\mu_\pm, X\,\text{var}_\pm\right). \tag{S2.7}$$

Also, according to [10] , the sum of two normal distributions is also a normal distribution, the mean and standard deviation of which are equal to the sums of the respective distribution's parameters. Therefore, the sum in Eq. (S2.3) will have the following distribution:

$$S(A,B) \to \text{norm}\left(A\mu_+ - B\mu_-, A\,\text{var}_+ + B\,\text{var}_-\right). \tag{S2.8}$$

The SDF is now determined by the sum $S(A,B)$ and the average of each function $P(A,B)$ over recombination volume distribution (25):

$$P_{A+B>1}\left(\text{Sign}\cdot R^2\right) = \sum_{A+B>1}\left\{ S(A,B)\int_{r'=0}^{\infty} P\left\{A,B,\frac{4}{3}\pi r'^3\cdot Vn_0\cdot\gamma\right\}\rho'\left(r'\right)dr'\right\}. \tag{S2.9}$$

Finally, we substitute $P(a,b)$ from Eq. (S1.4) and average over the distribution of volumes:

$$P = P_0 + P_{A+B>1}. \tag{S2.10}$$

The integral over r' is reduced to an integral with two parameters V and the sum $(A+B)$:

$$\begin{aligned}
&\int_{r'=0}^{\infty} P\left\{A,B,\frac{4}{3}\pi r'^3\cdot Vn_0\cdot\gamma\right\}\rho'\left(r'\right)dr' = \\
&= \left\{2\pi\sqrt{AB}\left(A/e\right)^A\left(B/e\right)^B\right\}^{-1}\left(\frac{1+\Delta}{2}\right)^A\left(\frac{1-\Delta}{2}\right)^B\times \\
&\times\left(\frac{4}{3}\pi\cdot Vn_0\cdot\gamma\right)^{A+B}\times\int_{r'=0}^{\infty}\left(r'^3\right)^{A+B}\exp(-\frac{4}{3}\pi r'^3\cdot Vn_0\cdot\gamma)\rho'\left(r'\right)dr'.
\end{aligned} \tag{S2.11}$$

Here Stirling's approximate formula for factorial is used:

$$n! \approx \sqrt{2\pi n}\left(n/e\right)^n. \tag{S2.12}$$

The integral becomes:

$$I = \int_{r'=0}^{\infty}\exp\left(3\left(a+b\right)\log\left(r'\right) + D + Er' + Cr'^2 - \frac{4}{3}\pi\cdot Vn_0\cdot\gamma r'^3\right)dr'. \tag{S2.13}$$

The integral is taken using the saddle-point method:

$$I \approx 1.4\times\exp(f)\left(-f''/2\right)^{-1/2}\frac{\sqrt{\pi}}{2}\text{erfc}\left(\left(-\tilde{r}\right)\sqrt{-f''/2}\right). \tag{S2.14}$$

The factor 1.4 is chosen to correct for the approximate nature of the saddle point method. Here the parameterized values of $\tilde{r}$, f and f'' are:

$$\tilde{r}=\left(8\pi Vn_0\right)^{-1}\left\{\begin{array}{l}\left(-0.0777\left(Vn_0\right)^{0.866}+0.02\right)(A+B)+\\ +5.66\left(Vn_0\right)^{0.78}(A+B)^{1/2}+11.75\left(Vn_0\right)^{0.75}-1\end{array}\right\}. \tag{S2.15}$$

$$f''=-3(A+B)/\tilde{r}^2-30-\left(16\pi\left(Vn_0\right)/3\right)\tilde{r}\,. \tag{S2.16}$$

$$f=3(A+B)\log\left(\tilde{r}\right)-15\cdot 0.9^2/2+20\tilde{r}-15\tilde{r}^2-\left(8\pi\left(Vn_0\right)/9\right)\tilde{r}^3\,. \tag{S2.17}$$

In the range Vn_0 from 2 to 30 and $(A+B)<100$ the above approximation error is verified to be less than 20 %. For small values of $(1< Vn_0<2)$, the main contribution to the distribution comes from the initial distribution P_0. The region of small Vn_0 was not used for fitting, since the correlation function comes to a stationary state at $Vn_0>2$ and, in addition, the application of the Central Limit Theorem in Eqs. (S2.7) and (S2.8) is reasonable only for large numbers A and B, i. e. for large Vn_0.

S3. Verification of the analytical versus numerical model

Here, analytic expressions developed in the main text for density kinetics and analytical SDF developed in Sections S2 and S3 are verified against Monte-Carlo simulations. Calculations was carried out with a fixed parameter $\kappa = 1.57\cdot 10^4$ c^{-1}. Formally, the value κ corresponds to the parameters of zirconium at a temperature of 400°C with the radius of the loops $R_0 = 5$ nm, Burgers vector $b = 0.32$ nm.

In applying the numerical model for the SDF, the value of the relative decrease in number $n_0 n^{-1}$ is used as an equivalent of time to find the Vn_0 parameter of SDF via Eqs. (S1.9) and (S1.10). The analytical SDF (S1.11) for the case of initially equally sized loops is plotted as lines on the Figs. S3.1, S3.2 and S3.4. Analytic lines in the Figs. S3.3 and S3.5 for number density kinetics are Eqs.(25) – (26) for "correlated" and the same equations with substitution pointed in Eq. (31) for "uncorrelated" initial distribution. The calculation result at $\Delta = 0$ for the SDF at different moments is shown in Fig. S3.1 , negative sizes stand for loops of a negative sign.

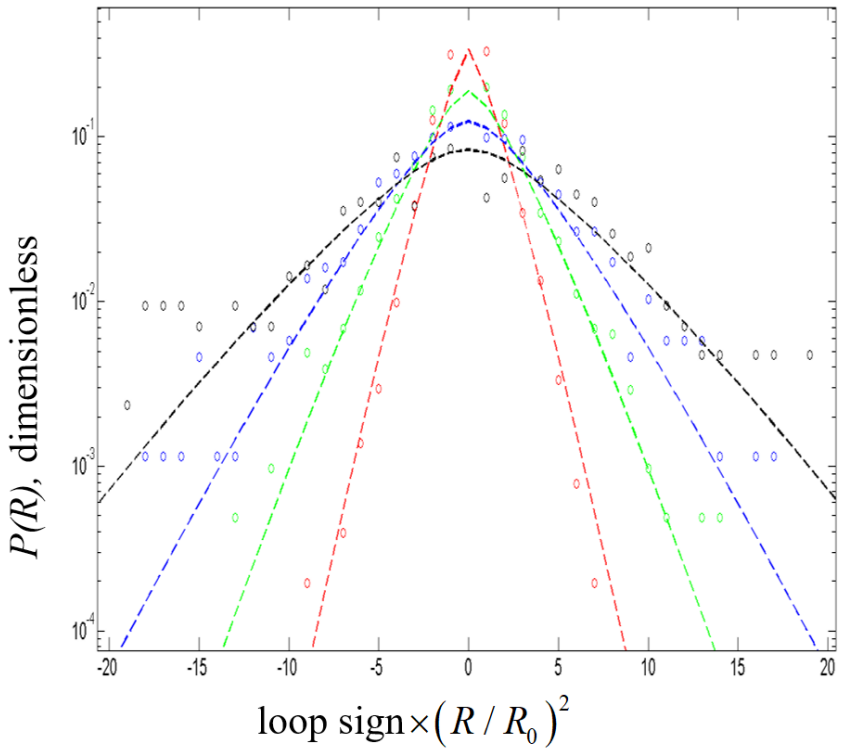

Fig. S3.1. (Color online) SDF distribution for initial $\Delta = 0$. The negative sign in the area is for negative loops. Colors for $n_0 n^{-1} = 3.0$; 7.2; 15.55; 33.3. Lines — model.

Results for SDF at initial $\Delta = 0.33$ are shown in Fig. S3.2, the loop density kinetics are shown in Fig. S3.3.

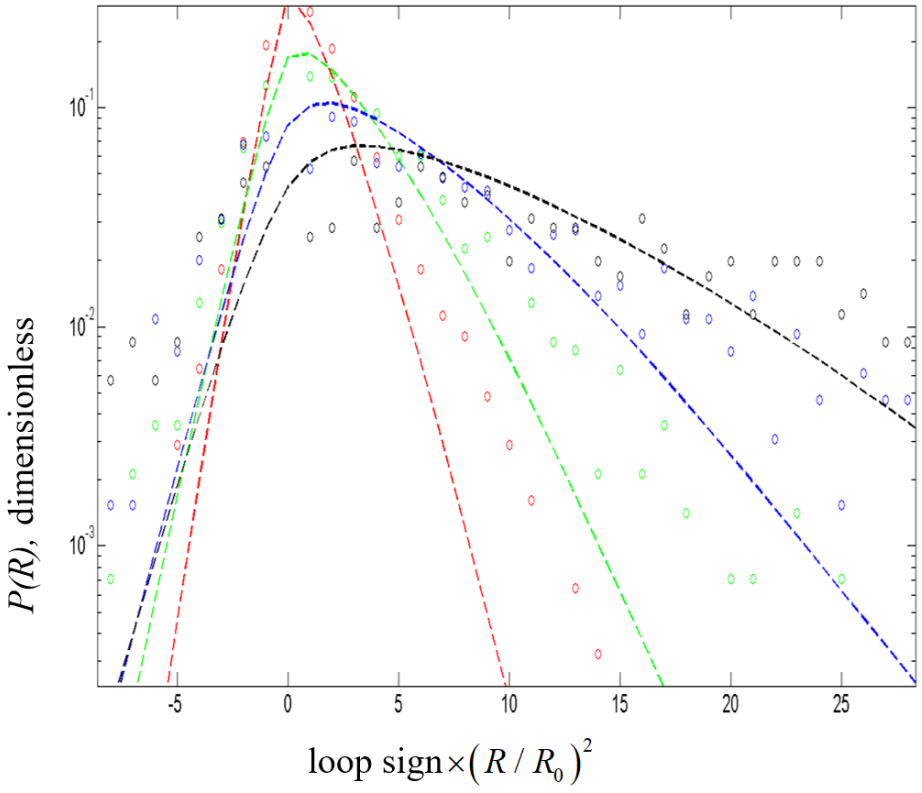

Fig. S3.2 (Color online) SDF for initial $\Delta = 0.33$. Colors for $n_0 n^{-1}$ = 3.0; 6.4; 13.9; 25.3. Dots — Monte-Carlo results, lines — analytical model.

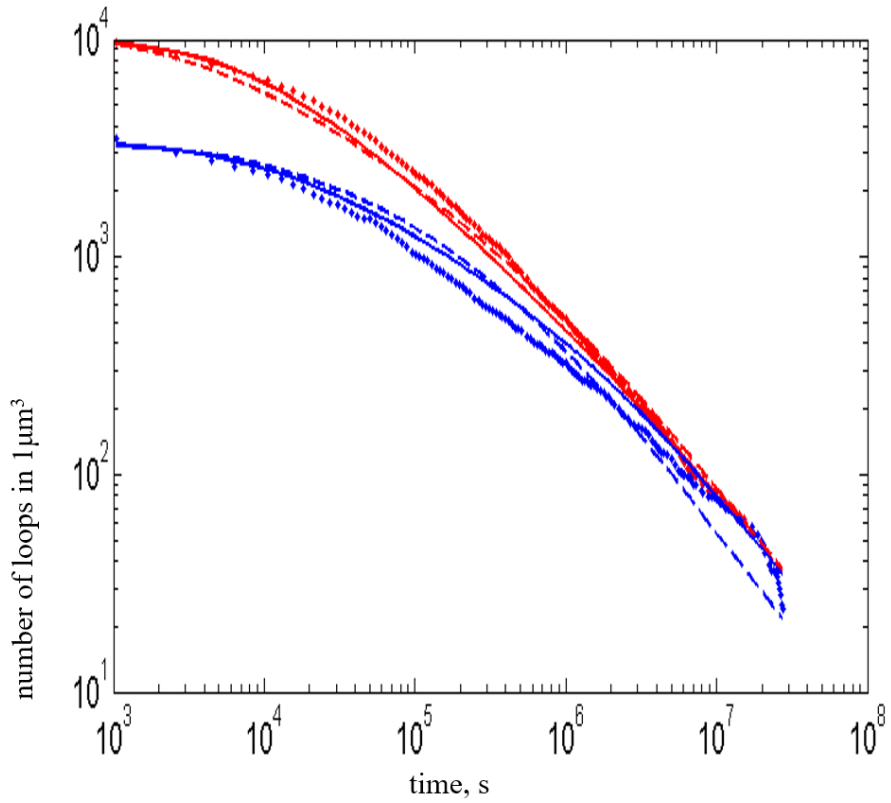

Fig. S3.3 (Color online) Kinetics for positive (red lines) and negative (blue lines) loops at $\Delta = 0.33$, $\kappa = 1.57 \cdot 10^4$ c^{-1}. Dots — Monte-Carlo results, the solid line is the analytic model for correlated distribution, the dashed line is the same for uncorrelated distribution.

https://doi.org/10.48612/letters/2025-4-297-303

The calculation result with initial $\Delta = 0.66$ for the SDF is shown in Fig. S3.4 and the kinetics is shown in Fig. S3.5.

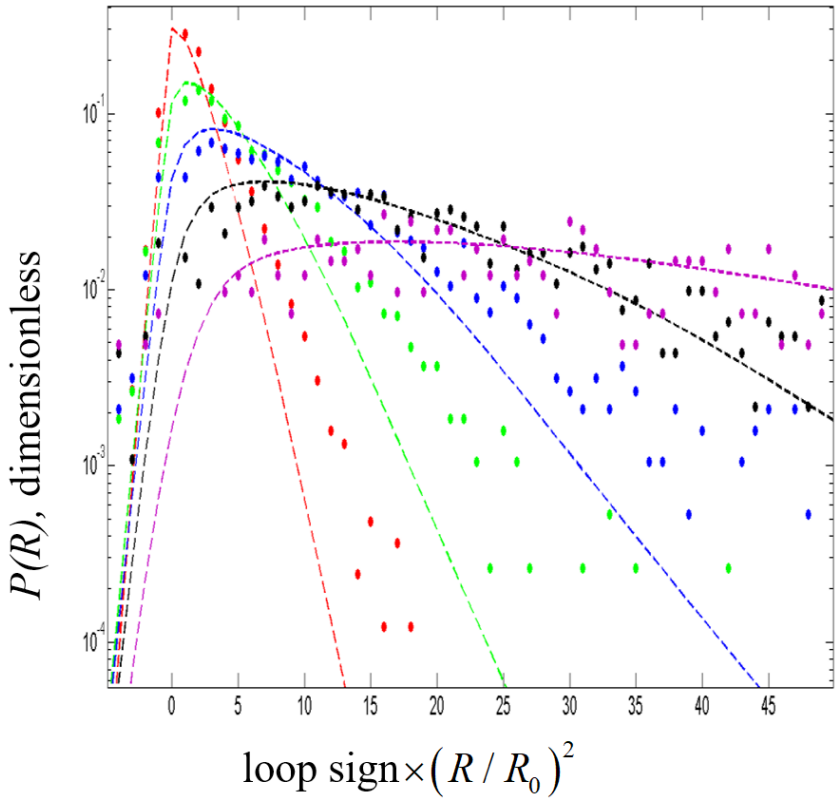

Fig. S3.4 (Color online) SDF for $\Delta = 0.66$, $\kappa = 1.57 \cdot 10^4$ c^{-1}. Dots — Monte-Carlo results, lines — analytical model. The negative sign is for negative loops. Colors for $n_0 n^{-1} = 2.86$; 6.2; 12.4; 25.6; 57.1.

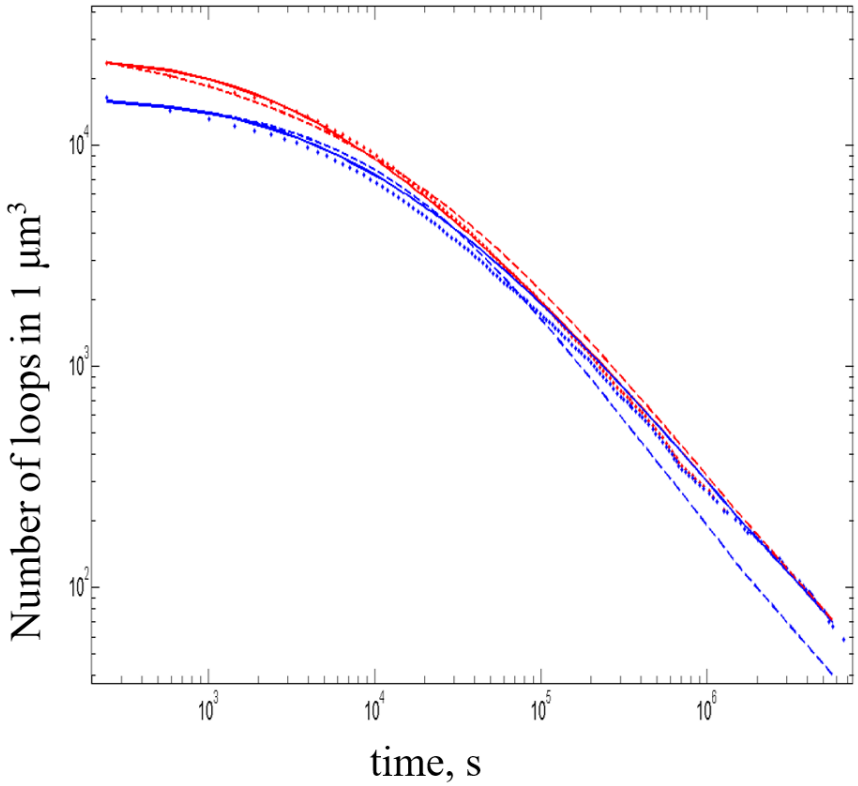

Fig. S3.5 (Color online) Kinetics for positive (red) and negative (blue) loop numbers with initial $\Delta = 0.66$, $\kappa = 1.57 \cdot 10^4$ c^{-1}. Dots — Monte-Carlo results, solid and dashed lines — calculation in correlated and uncorrelated distribution.

Finally, Fig. S3.6 shows analytic and numerical calculations with an initially continuous distribution (red line), represented by two different normal distributions for positive and negative loops. As in the previous cases of unimodal loop size $R = \delta(R_0)$, the analytical model (S2.8), (S2.10), (S2.11) – (S2.14) is in good agreement with the calculated SDF for the predominant loop type, though underestimating the size growth of the minor type.

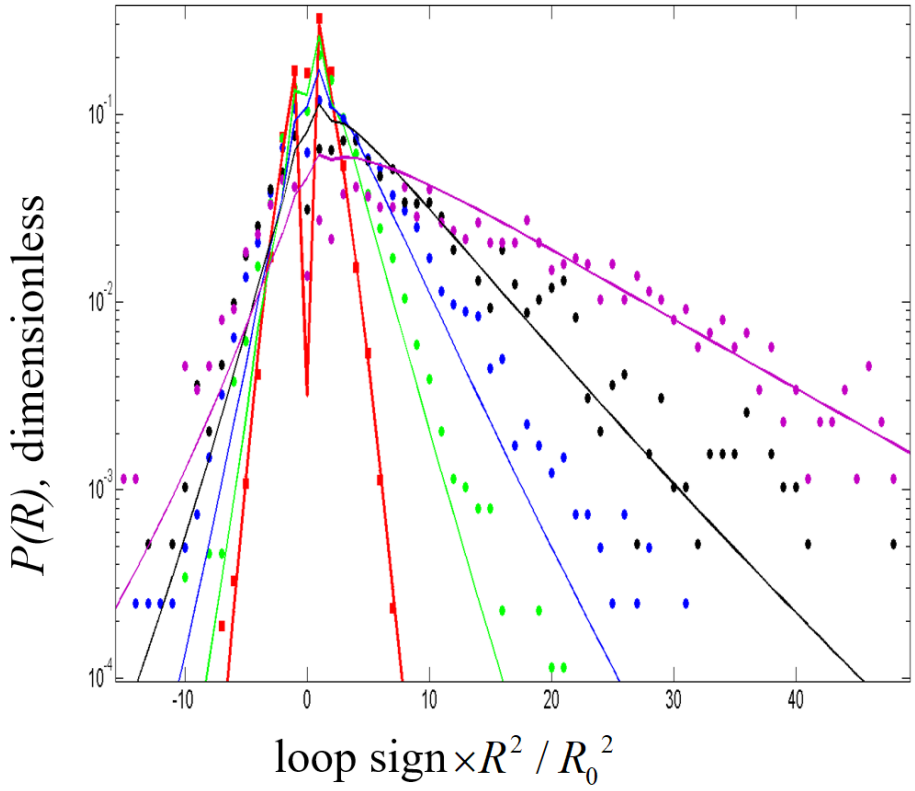

Fig. S3.6 (Color online) SDF for initial parameters: $\Delta = 0.33$, normal distribution for both loop types with $\mu_+ = 1.2R_0^2$, $var_+ = 0.2R_0^4$ for positive and $\mu_- = 1.2R_0^2$, $var_- = 0.16R_0^4$ for negative. Dots — Monte-Carlo results, line — analytic model (S2.9), (S2.10), (S2.11), (S2.14). Colors for $n_0 n^{-1}$: 1.0; 2.8; 6.04; 12.6; 27.9.

https://doi.org/10.48612/letters/2025-4-297-303