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Mechanism of internal friction in bulk $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ metallic glass

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A mechanism of internal friction in metallic glasses is developed and adopted to analyze the results measured in the bulk $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ metallic glass. A kink of internal friction is observed near the calorimetric glass transition. The nonlinear relation between the internal friction and frequencies is observed. The obtained activation energies for the relaxation in glassy state (1.7 eV) and supercooled liquid state (5.2 eV) suggest that the relaxation should be mainly controlled by the motion of a single atom in the glassy state, while by collective motion of atoms in the supercooled liquid state. The nonlinear relation between the internal friction and frequencies should be mainly caused by the distribution of microscopic relaxation parameters. The effects of annealing on relaxation parameters are also observed and discussed.

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I. INTRODUCTION

The success of the internal friction (IF) method in the studies of the relaxation of point defects, dislocations, grain boundaries, etc., shows that it is a very useful tool to explore the relaxation processes in metals and alloys.[1] Hence, it was widely used to study the relaxation in both the glassy state (GS) and supercooled liquid state (SLS) of the metallic glassy ribbons, and some interesting results were obtained.[2,3] After it is possible to produce bulk metallic glasses (BMGs) at critical cooling rates as low as 1 K/s,[4,5] the IF method has also been used to study the relaxation of BMGs. Among these works, some were concentrated on the hydrogen-induced IF or damping peak,[6,7] while others emphasized on the intrinsic structural relaxation of BMGs.[8–10] In addition, the α relaxation associated with the large-scale molecular movements and the β relaxation associated with the localized molecular motion below the temperature of glass transition T_g were usually observed in bulk Zr-Ti-based metallic glasses MGs.[11–13] As for the mechanism of internal friction or damping in MGs, most models given in the literature show that the IF $\tan\phi$ (ϕ is the angle by which the strain lags behind the stress) in the range of low frequencies can be approximately expressed as[1,10]

$$\tan\phi = G/A\omega\eta, \qquad (1)$$

where G is the elastic modulus, $\omega=2\pi f$, f is the measuring frequency, η is the effective viscosity, and A is a constant. Hence a linear relation between $\tan\phi$ and $1/f$ is expected. Although it was indeed observed in Co-based BMGs,[10] a nonlinear relation was observed in our studies. The frequency-dependence relation of internal friction is very important to understanding the mechanism of relaxation in BMGs. Unfortunately, very limited work on this aspect can be found up to now. Furthermore, the broad distribution of microscopic relaxation processes should be naturally expected in noncrystalline solids. However, the existed models fail to give any image on the distribution of relaxation parameters. Hence, a reconsideration of the mechanism of internal friction in BMGs is both necessary and important to explore the intrinsic relaxation in BMGs.

II. EXPERIMENTS AND RESULTS

The $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ BMG with dimensions of $40\times 2\times 50\ \mathrm{mm}^3$ were obtained by centrifugal casting technique. The amorphous structure was confirmed by x-ray diffraction (XRD), as shown in Fig. 1. The XRD patterns were recorded by means of a Philips PW1700 diffractometer using Cu Kα radiation. Differential scanning calorimetry (DSC) results re-

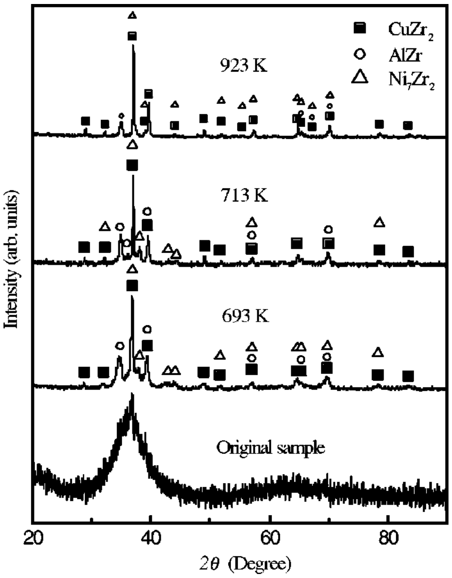

FIG. 1. XRD patterns for original $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ sample and annealed samples for 3600 s at 693, 713, and 923 K respectively.

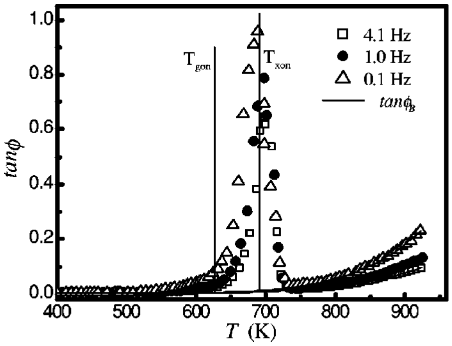

FIG. 2. Internal friction tanϕ as a function of temperature T for as-cast $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ BMG at a heating rate of 0.033 K/s.

vealed that the onset temperature of calorimetric glass transition T_{gon} and crystallization T_{xon} are 642 K and 734 K, respectively, at a heating rate of 0.33 K/s. Based on T_{gon} and T_{xon} at heating rates of 0.33, 0.67, 1.33, and 2.67 K/s, the activation energies were calculated to be 4.90 ± 0.19 eV for glass transition by the Arrhenius plot,[14] and 2.27 ± 0.13 eV for crystallization by the Kissinger equation. The apparatus for internal friction measurements is an automatic inverted torsion pendulum with the error of about 0.001. The IF was measured at 0.1, 0.3, 1.0, 2.0, and 4.0 Hz in a single heating run at a rate of 0.033 K/s. The strain amplitude is 2×10^{-5}.

Figure 2 shows the internal friction tanϕ for the as-cast $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ BMG. The calculated onset temperatures of calorimetric glass transition T_{gon} and crystallization T_{xon} at heating rates of 0.033 K/s are 625 K and 692 K, respectively, and also marked in the figure. A pronounced IF peak occurs at about 690 K, almost the same temperature of the T_{xon}. It suggests that the origin of the peak should be mainly caused by the beginning of crystallization. Similar results but with a much lower peak height were also observed for the sample annealed at 693 K for 3600 s. The peak completely disappears for the sample annealed at 713 K for 3600 s. The corresponding XRD patterns for the samples annealed at 693, 713, and 923 K are also shown in Fig. 1. It can be found that most XRD peaks can be identified by the tetragonal $CuZr_2$ phase (SG 139), and the remaining peaks can be identified as orthorhombic ZrAl (SG 63) and monoclinic Ni_7Zr_2 (SG 12) phases. Nevertheless, the pattern for the sample annealed at 713 K closer to that of the sample annealed at 923 K. The combined XRD and IF results indicate that the sample annealed at 693 K should be partially crystallized.

One can see from Fig. 2 that the internal friction peak is superimposed by a background tanϕ_B, which can be generally expressed as[1]

$$\tan\phi_B = B + Ce^{-Q_b/kT}, \tag{2}$$

where both B and C are frequency-dependent constants, Q_b is the apparent activation energy, and T is the absolute temperature. Using a fitting program, the tanϕ_B for different f was obtained, as shown in Fig. 2 by solid lines. By subtracting tanϕ_B from the measured tanϕ, the internal friction associated with the peak tanϕ_P was obtained. Figure 3 shows the Arrhenius plots of tanϕ_P for both the as-cast sample and the sample annealed at 693 K.

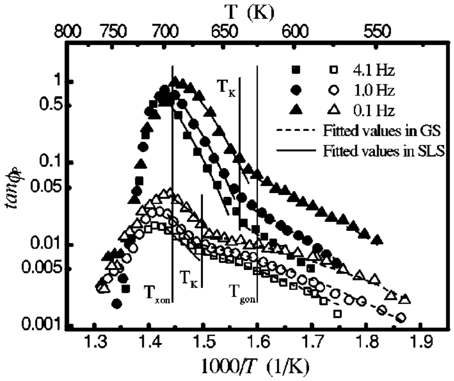

FIG. 3. Arrhenius plots of the tanϕ_P for as-cast (solid symbol) and annealed (open symbol) $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ BMG at a heating rate of 0.033 K/s, and the fitted results [Eq. (10)].

It can be clearly seen from Fig. 3 that a kink of tanϕ_P exists for both samples. The kink temperature T_K was occasionally regarded as an indication of the T_g. It is usually accepted that the relaxation below T_g was regarded as β relaxation, while that above T_g was thought to be α relaxation.[11,12] For the as-cast sample, the T_K is about 638 K, which is about 15 K higher than T_{gon} and close to the ending temperature of the glass transition; while for the annealed sample, T_K shifts to 668 K while T_p remains almost the same. As usual, the average activation energy Q_β (slope of lines) of different frequencies for the β relaxation is evaluated to be 0.80 ± 0.07 eV for the as-cast sample, which is close to that (0.7 eV) obtained in ZrTiCuNiBe BMGs.[11,12] However, the Q_β is only 0.44 ± 0.09 eV for the annealed sample. It is unreasonable that the localized atom moves with a smaller energy barrier in the denser packed atom structure. In addition, detailed analysis of tanϕ_P at the same temperature but at different frequencies f reveals that the linear relation of the tanϕ_P and $1/f$ does not exist. Instead, a nonlinear relation was found

$$\tan\phi_P \propto 1/f^n, \tag{3}$$

where n is the apparent nonlinear index. The average values of n for different temperatures change from 0.45 for the as-cast samples to 0.30 for the annealed samples, i.e., the index n should be a microstructure-sensitive parameter of BMGs. Hence, reconsideration of the mechanism of internal friction of BMGs is both necessary and important.

In order to study the effects of temperature on the structural relaxation, the internal friction of $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ BMG was measured during annealing at different temperatures. Figure 4 shows the variation of tanϕ during annealing. The results can be summarized as: (i) tanϕ keeps invariant at 653 K, (ii) tanϕ is invariant for the initial 1300 s and then decreases gradually at 673 K, (iii) tanϕ continuously decreases at 693 K ($\sim T_p$), and (iv) tanϕ is very small and displays small variations at 713 K.

For the sake of obtaining the intrinsic relation of tanϕ and f, only 653 K and 713 K were chosen to measure the tanϕ $\sim f$ spectrum. The results are shown in Fig. 5. It should be pointed out that the data in Fig. 5 were averaged over three

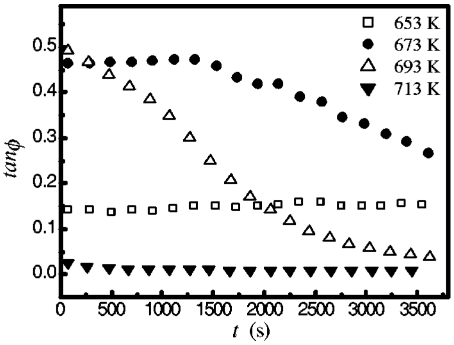

FIG. 4. Internal friction $\tan\phi$ as a function of time during annealing at different temperatures; the measuring frequency is 1.0 Hz.

measurements. The period is about 1200 s for a whole measurement. Hence, for the same frequency, the time at the third measurement is about 2400 s later than that at the first one. The small error of the $\tan\phi$ also suggests that the annealing on these two temperatures has little influence on the $\tan\phi$, i.e., the obtained relation between $\tan\phi$ and f should be an intrinsic property of the bulk glassy $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ alloy. One can find that an approximately linear relation (dotted line) between $\ln(\tan\phi)$ and $\ln f$ exists at 653 K. The nonlinear index n was evaluated to be 0.45 from the slope, the same value evaluated from the internal friction-temperature $\tan\phi_P \sim T$ spectrum. Reminding that $\tan\phi$ includes the contribution of the background $\tan\phi_B$. However, the $\tan\phi_B$ is small and increases slowly with temperatures in both the GS and SLS (Fig. 2). Hence, the influence of the background $\tan\phi_B$ on the obtained relation is small. As for the internal friction at 713 K, the level of it is small. It should be mainly consisted of the background $\tan\phi_B$.

III. MECHANISM

As mentioned previously the existed model on the internal friction $\tan\phi$ of metallic glasses [Eq. (1)] cannot give a satisfied explanation of the present results, especially the nonlinear relation between $\tan\phi$ and frequency f in the present range of low frequencies. Accordingly, a mechanism of the internal friction in metallic glasses is to be developed in the following.

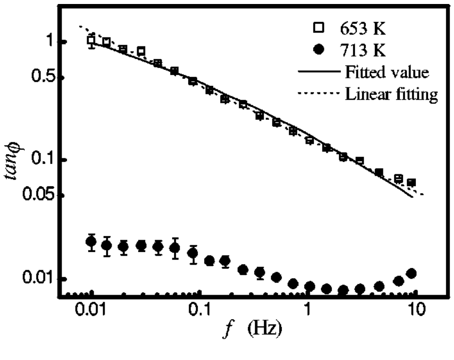

FIG. 5. Double logarithmic plots of internal friction $\tan\phi$ vs frequency f at different temperatures, and the fitted result [Eq. (10)].

Under the action of the alternating stress σ_a $(=\sigma_0 e^{i\omega t}$, σ_0 the stress amplitude and t is the time) for internal friction measurements, the total strain should be the sum of all viscoelastic strain ε_{vi} caused by the motion of the ith relaxation center (single atom or atom clusters) and the homogeneous elastic strain ε_u $(=\sigma_a/G=\sigma_a J_u$, J_u is the unrelaxed compliance). Assuming the motion of each relaxation center can be represented by a Voigt model,[1] the ε_{vi} satisfies

$$\eta_i\dot{\varepsilon}_{vi} + K_i\varepsilon_{vi} = \sigma_a, \tag{4}$$

where K_i is the restoring force representing the action of the neighboring atoms on the relaxation center. From Eq. (4), one can easily obtain the complex compliance

$$J_{vi} = \frac{\varepsilon_{vi}}{\sigma_a} = \frac{1}{K_i + i\,\eta_i\omega}. \tag{5}$$

By setting $K_i=0$ and dividing J_{vi} by the unrelaxed compliance J_u, one can immediately obtain Eq. (1), i.e., the result of Maxwell model.[1,10] In fact, the condition that $K_i=0$ should be valid only for fluids. According to Eq. (5), the storage and loss compliances J_{vi1} and J_{vi2} are, respectively,

$$J_{vi1} = \frac{\delta J_i}{1+\omega^2\tau_i^2}, \tag{6a}$$

$$J_{vi2} = \delta J_i\frac{\omega\tau_i}{1+\omega^2\tau_i^2}, \tag{6b}$$

where $\delta J_i(=1/K_i)$ is the relaxation of the compliance and the relaxation time τ_I is

$$\tau_i = \eta_i/K_i. \tag{7}$$

Due to the existence of lots of such relaxation centers, a continuous distribution of the relaxation time τ should exist. In general, a Gaussian distribution function is assumed[1]

$$X(z) = \delta J\beta_o^{-1}\pi^{-1/2}e^{-(z/\beta)^2}, \tag{8}$$

where $z=ln(\tau/\tau_m)$. τ_m is the mean relaxation time, δJ is the total relaxation of the compliance and β_o is a measure of the width of the distribution. Replacing δJ_i in Eqs. (6)(a) and (6)(b) by $X(z)dz$, and the integration yields

$$J_{v1} = J_1 - J_u = \frac{\delta J}{\sqrt{\pi}}\int_{-\infty}^{+\infty}\frac{e^{-w^2}}{1+e^{2(x+\beta_o w)}}dw, \tag{9a}$$

$$J_{v2} = J_2 = \frac{\delta J}{\sqrt{\pi}}\int_{-\infty}^{+\infty}\frac{e^{-w^2}}{e^{x+\beta_o w}+e^{-(x+\beta_o w)}}dw, \tag{9b}$$

where $x=ln(\omega\tau_m)$, $w(=z/\beta_o)$ is the integrating variable, and J_1 and J_2 are the total storage and loss compliance, respectively. According to the definition of internal friction, one can obtain

TABLE I. Relaxation parameters for GS and SLS in $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ BMG.

	Glassy state				Supercooled liquid state			
	Δ	Q_m: eV	τ_{m0}: s	β_o	Δ	Q_m: eV	τ_{m0}: s	β_o
As-cast	3.88	1.69	1.9×10^{-9}	7.4	16.6	5.18	3.5×10^{-36}	6.4
	$\tan\phi\sim f$ spectrum			17.4	51.3	3.8×10^{-36}	6.2	
Annealed	0.21	2.09	6.8×10^{-14}	9.9	1.05	4.29	6.2×10^{-28}	8.4

$$\tan\phi = \frac{J_2}{J_1} \approx \frac{\Delta}{\sqrt{\pi}}\int_{-\infty}^{+\infty} \frac{e^{-w^2}}{e^{x+\beta_o w} + e^{-(x+\beta_o w)}}dw, \qquad (10)$$

where $\Delta(\approx \delta J/J_u)$ is the relaxation strength.

IV. DISCUSSION

Before using the Eq. (10), the expression of the mean relaxation time τ_m on temperatures must first be known. According to Eq. (7), the temperature dependence relation of τ_m should be mainly determined by the effective viscosity η_i due to the relatively slow change of the restoring force constant K_i. According to Angell's classification, the η_i and τ_m of the strong liquids behave in nearly Arrhenius fashion, while fragile liquids show Vogel-Fulcher-Tammann (VFT) relation.[15,16] It is generally thought that the BMGs are rather "strong" compared to the fragile glasses relative to the conventional MGs. Zapple and Sommer also found that the η_i of the present alloy could be described by an Arrhenius relation.[14] Hence the mean relaxation time τ_m is can be expressed as

$$\tau_m = \tau_{m0}e^{Q_m/kT}, \qquad (11)$$

where τ_{m0} and Q_m are the pre-factor and activation energy of τ_m respectively. It should be noted that the present mechanism may be extended to fragile glasses if we replace Eq. (11) with the VFT relation.

According to Eq. (10), a fitting program was developed based on the Bevington method.[17] Due to the existence of the kink, the $\tan\phi_P$ was divided into two subsets according to the kink temperature T_K. By fitting the $\tan\phi_P$ of different T and f together, the relaxation parameters in GS and SLS for the as-cast sample were obtained, respectively, as shown in Table I. Similar results were obtained from the internal friction-frequency $\tan\phi\sim f$ spectrum. Using the same method, the corresponding relaxation parameters for annealed samples were also obtained, and shown in Table I. The fitting results were also shown in Figs. 3 and 5 by lines. One can find that there is good agreement between the fitted and experimental results. Thus, we have obtained the activation energy and preexponential factor of relaxation time for both glassy state and supercooled liquid state by internal friction measurements. Now we can clearly understand that the observed nonlinear relation between the internal friction $\tan\phi$ and frequencies f should be caused by the distribution of microscopic relaxation processes. The change of the distribution parameters will result in the change of the value of the apparent nonlinear index n [Eq. (3)].

From Table I, one can find that the mean activation energy for the relaxation Q_m (5.2 eV) in supercooled liquid state is much larger than that (1.7 eV) in glassy state for the as-cast BMG. It is striking to be noted that the difference of the magnitude of the τ_{m0} in GS ($\sim10^{-9}$ s) and SLS ($\sim10^{-36}$ s) is over 27 orders. Similar results have also been observed in the study on the diffusion coefficient D of Be in ZrTiCuNiBe BMGs.[18–20] As mentioned previously, the Q_m should be mainly associated with the effective viscosity η. According to the Stokes-Einstein relation,[16,19]

$$D = kT/(6\pi\eta R), \qquad (12)$$

where D is the diffusion coefficient, R is the atomic radius. Hence an inverse relation exists for the D and η. Although Geyer and his collaborator[19] found that the exact Stokes-Einstein relation does not hold for ZrTiCuNiBe BMGs, the diffusion coefficient obtained from viscosity and measured data have a similar temperature-dependence relation and the difference of them decreases with increasing temperatures. In addition, the measured viscosity η should be associated with slower constituents because the viscous deformation is mainly governed by the mobility of slower constituents (generally larger atoms). Accordingly, the activation energy obtained here should be associated with that of the diffusion of larger atoms.

As for the reason for the marked difference between Q_m and τ_{m0} of diffusion coefficients in GS and SLS, there are detailed discussions in Refs. 19 and 20. The small isotope effect of diffusion observed in the study of the mass dependence of diffusion suggested that the collective hopping of atoms governs long-range diffusion in SLS. Furthermore, the hopping is a highly cooperative process involving about ten atoms.[21,22] Tang *et al.*[23] clearly pointed out that the single-atom hopping is an intrinsic property of both the relaxed GS and SLS, while the collective motion of atoms is the dominant process for the diffusion in SLS. The present results also show that the relaxation in $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ BMG should be mainly controlled by the viscoelastic motion of single atoms in the glassy state, while by the collective motion of atoms in the supercooled liquid state.

It is worthwhile to noted that the obtained distribution parameter $\beta_o(6\sim10)$ of relaxation time τ_m for the present BMG is much larger than the $(3\sim4)$ observed in crystalline[1,24] and nanocrystalline materials.[25] This result indicates that the relaxation should be controlled by the processes with a very broad distribution of τ_m. One can find from Eq. (7) that the τ_m is determined by both the effective viscosity η_i and the restoring force constant K_i, which were mainly determined by the atomic structure of relaxation centers and the arrangement of neighboring atoms. Comparing to crystalline materials, the BMGs have very complex atomic structure and configuration. Hence, a broader distribution of τ_m should naturally be expected.

For the sample annealed at 693 K, the marked decrease of Δ suggests that the BMG should be partially crystallized. The increase of Q_m and decrease of τ_{m0} in GS should be caused by the stronger interaction between atoms resulted from the increasing density, which is induced by the partial crystallization, while the decrease of the Q_m and the increase of τ_{m0} in SLS should be associated with the decreasing size of collective moving atom clusters because the larger ones are more difficult to move in the partially crystallized structure. It is interesting to note that the τ_{m0} in GS ($\sim 10^{-14}$ s) for the annealed samples is of the same order of magnitude of that for crystalline materials (the reciprocal of Debye frequency).[1,24] This is also an indication that the relaxation in glassy state should be controlled by the motion of a single atoms.

As for the decrease of the internal friction $\tan\phi$ during annealing at temperatures higher than 673 K, it is mainly caused by the irreversible structure relaxation and the beginning of the crystallization. But it is surprising that the $\tan\phi$ keeps almost invariant at 653 K because the free volume reduction Δv should apparently occur at this temperature. Investigation on the correlation between enthalpy change and Δv of ZrCuAlNi BMG[18] showed that the low-temperature structure relaxation should be also connected with topological short-range ordering (TSRO) and not only with chemical short-range ordering (CSRO). As for the reason for the invariance of $\tan\phi$ at 653 K, we think that the change of $\tan\phi$ induced by Δv may be much smaller than that caused by the intrinsic viscoelastic relaxation. In addition, Δv caused by the TSRO and CSRO may already be completed during the heating to the annealing T with relatively slow heating rate (~ 0.17 K/s). Similar results were also observed by the comparative internal friction study of bulk and ribbon glassy ZrTiCuNiAl alloys.[8]

V. CONCLUSION

A mechanism of internal friction in metallic glasses was developed and adopted to analyze the results measured in $Zr_{65}Cu_{17.5}Ni_{10}Al_{7.5}$ BMG. An apparent kink of internal friction exists near the calorimetric glass transition. The nonlinear relation between the internal friction and frequencies was observed. According to the derived expression, a fitting program was developed to obtain the relaxation parameters. The obtained activation energy for the relaxation (1.7 eV) in glassy state and (5.2 eV) in supercooled liquid state suggest that the relaxation should be mainly controlled by the motion of single atoms in glassy state, and by the collective motion of atoms in the supercooled liquid state. The nonlinear relation between the internal friction and frequencies should be mainly caused by the distribution of microscopic relaxation parameters. The effects of annealing on the relaxation parameters were also observed, and the annealing will result in the decrease of the size of collective moving atoms. Finally, very broad distributions of relaxation times were found to be existed in both the as-cast and annealed samples.

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