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Waiting time dependence of aging

Takashi Odagaki

Kyushu University, Fukuoka 819-0395, Japan
Research Institute for Science Education Inc, Kyoto 603-8346, Japan

E-mail: t.odagaki@kb4.so-net.ne.jp

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Abstract

Aging phenomena have been observed in many non-equilibrium systems such as polymers and glasses, where physical properties depend on the waiting time between the starting time of observation and the time when the temperature is changed. The aging is classified into two types on the basis of the waiting time dependence of an instantaneous relaxation time: When the relaxation time is always an increasing function of the waiting time, the aging is called Type I and when it depends on the protocol of the temperature change, the aging is called Type II. Aging of a random walk in three dimensions is investigated when the free energy landscape controlling the jump rate responds to temperature change with a delay. It is shown that the intermediate scattering function of the random walk model exhibits Type II aging. It is also shown that the relaxation time of the free energy landscape can be deduced from the waiting time dependence of the instantaneous relaxation time.

Keywords: non-equilibrium system, aging, waiting time dependence, free energy landscape, instantaneous relaxation time

(Some figures may appear in colour only in the online journal)

1. Introduction

Many systems are known to stay in a non-equilibrium state for a long time due to the separation of slow dynamics and fast dynamics, which include glass forming materials, high polymers and spin glasses. In the non-equilibrium systems, physical properties show delayed response to a sudden change in temperature and depend on the observation time. This phenomenon is called aging [1–5]. Conventionally, the aging is understood by the time required for the probability distribution of microstates to be re-established and is represented by a relaxation function. Well-known relaxation functions are the exponential function $\exp(-t/\tau)$ (the Debye relaxation) and the stretched exponential function $\exp\left\{-(t/\tau)^{\beta}\right\}$ (the Kohlrausch–Williams–Watts or KWW relaxation) where $\beta < 1$ and τ are positive constants supposed to be intrinsic parameters of the system. The difference between these functions is in their waiting time dependence. Namely, when the temperature is changed at time $t = 0$ and the observation is started after a waiting time t_w, the relaxation time of the KWW relaxation is an increasing function of the waiting time. Some of experimental data for relaxation can be fitted by a power-law function [6]. The instantaneous relaxation time of the power-law relaxation defined at a waiting time is also an increasing function of the waiting time. The Debye relaxation does not show the waiting time dependence.

It has been argued that trap models with a power law distribution of jump rates give rise to the slow relaxation following the KWW relaxation [7, 8] or a power-law decay [8] of the relaxation function.

Kob and Barrat [9] investigated by molecular dynamics simulation the waiting time dependence of the intermediate scattering function for a binary Lennard-Jones liquid in the super-cooled region, when the system was quenched at $t = 0$ (T-down protocol) and the observation was started after a waiting time t_w. They showed that the intermediate scattering function depends strongly on the waiting time and the structural relaxation time becomes longer as the waiting time is increased. This behavior has been considered to be the essence of slow dynamics in which the relaxation time is an increasing function of the waiting time.

Suarez *et al* [10] investigated out-of-equilibrium dynamics of a fractal model gel by molecular dynamics simulation when the temperature is raised from zero (T-up protocol). They reported that when the temperature is low, the dynamics slows down with increasing waiting time and that when the temperature rise is large, the relaxation becomes faster with increasing waiting time, which is attributed to the waiting time dependence of exponent β of the stretched exponential relaxation function due to breaking of the fractal network.

These two computer simulations raise an important question if the waiting time dependence of aging depends on the protocol of temperature control, and if it does it is important to find out physics behind the waiting time dependence of aging.

Conflicting protocol dependences of aging have been reported for spin glasses. On one hand, Granberg *et al* showed that the T-drop experiment requires a longer waiting time to establish the same correlation length than the T-up experiment [11]. On the other hand, Lederman *et al* showed that the T-up and T-down experiments yield the same waiting time dependence of the relaxation of the thermo-remanent magnetization [12].

For structural glasses, the protocol dependence has been found for the volume and mechanical behavior of the epoxy [13] and for the dielectric relaxation of a glass-forming liquid [14].

The slow structural relaxation in the non-equilibrium systems can be described by the time evolution of a representative point on the free energy landscape (FEL) determined by the fast motion [15]. In the FEL approach to non-equilibrium systems, the FEL is defined as a function of temperature, and when the temperature is modified, the FEL will be re-established with time delay. In fact, it has been shown that such time dependence of the FEL can explain the observation in the temperature-modulation spectroscopy [16, 17].

In this paper, I first discuss general properties of a relaxation function, a two-time relaxation function and show that the aging can be classified into two types, Type I and Type II, on the basis of the protocol dependence of the instantaneous relaxation time. I investigate how the delayed response of the FEL to temperature change manifests itself in the waiting time dependence of aging. As a model system, I study the intermediate scattering function of a random walk model in three dimensions and show that the delayed response of the FEL gives rise to Type II aging. I also show that the relaxation time of the FEL can be deduced from the waiting time dependence of aging.

I organize this paper as follows. I first explain basic concepts including aging and definitions of relevant quantities in section 2. I also define Type I and Type II agings, where the latter is characterized by the protocol dependence of aging. In section 3, the free energy landscape approach is explained briefly and a model for delayed response of the FEL to a temperature change is introduced. Exploiting the formalism, I investigate the intermediate scattering function for a random walk model in three dimensions in section 4. Section 5 is devoted to discussion.

2. Conventional description of relaxation and aging

I consider a temperature change:

$$T = \begin{cases} T_0 \ (t < 0) \\ T_1 \ (t \geqslant 0), \end{cases} \tag{1}$$

where $T_0 > T_1$ and $T_0 < T_1$ denote the T-down and the T-up protocol, respectively.

When the temperature follows equation (1), a physical quantity $\mathcal{F}(t)$ relaxes from an equilibrium value $\mathcal{F}(0) = \mathcal{F}_0$ for $t < 0$ to a new equilibrium value $\mathcal{F}(\infty) = \mathcal{F}_1$ in the limit of $t = \infty$. The time dependence of the physical quantity $\mathcal{F}(t)$ is conventionally written as:

$$\mathcal{F}(t) = \mathcal{F}_1 + (\mathcal{F}_0 - \mathcal{F}_1)\Psi(t), \tag{2}$$

where $\Psi(t) = [\mathcal{F}(t) - \mathcal{F}_1]/[\mathcal{F}_0 - \mathcal{F}_1]$, which satisfies $\Psi(0) = -1$ and $\Psi(\infty) = 0$, is called the relaxation function.

Using the relaxation function $\Psi(t)$, I define a relaxation exponent function $\psi(t)$ by:

$$\psi(t) = -\ln\Psi(t) \tag{3}$$

and an instantaneous relaxation time $\tau(t)$ by:

$$\tau(t) = \left[\frac{\mathrm{d}\psi(t)}{\mathrm{d}t}\right]^{-1}. \tag{4}$$

When $\Psi(t)$ and $\psi(t)$ depend on time, the relaxation is said to show aging. Exploiting the time dependence of the instantaneous relaxation time $\tau(t)$, I classify the aging into two types, Type I and Type II: when the sign of $\mathrm{d}\tau(t)/\mathrm{d}t$ does not depend on the protocol of temperature change, the aging is called Type I, and when it depends on the protocol, the aging is called Type II.

In aging experiments, an observation is often started at a waiting time t_w when the temperature is changed at time $t = 0$ and the physical quantity $\mathcal{F}(t_w + t')$ at $t = t_w + t'$ is analyzed as a function of t_w and t'. Then, a two-time relaxation function $\Psi(t', t_w)$ can be defined by:

$$\mathcal{F}(t_w + t') = \mathcal{F}_1 + (\mathcal{F}(t_w) - \mathcal{F}_1)\Psi(t', t_w), \tag{5}$$

or:

$$\Psi(t', t_w) = \frac{\mathcal{F}(t_w + t') - \mathcal{F}_1}{\mathcal{F}(t_w) - \mathcal{F}_1}, \tag{6}$$

and it can be shown from equation (2) that:

$$\Psi\left(t',t_w\right)=\frac{\Psi\left(t_w+t'\right)}{\Psi\left(t_w\right)} \tag{7}$$

which satisfies $\Psi\left(0,t_w\right)=1$ and $\Psi\left(\infty,t_w\right)=0$. Namely, the two-time relaxation function $\Psi\left(t',t_w\right)$ is related to $\Psi\left(t\right)$ at $t=t_w$ and $t=t_w+t'$.

I define a two-time relaxation exponent function $\psi\left(t',t_w\right)$ and a two-time instantaneous relaxation time $\tau\left(t',t_w\right)$:

$$\psi\left(t',t_w\right)=-\ln\Psi\left(t',t_w\right), \tag{8}$$

$$\tau\left(t',t_w\right)=\left[\frac{\mathrm{d}\psi\left(t',t_w\right)}{\mathrm{d}t'}\right]^{-1}. \tag{9}$$

From equation (7), it can be shown that:

$$\tau\left(t',t_w\right)=\tau\left(t_w+t'\right) \tag{10}$$

holds. Therefore, the type of aging can be identified by the protocol dependence of $d\tau\left(0,t_w\right)/dt_w$ as well.

As an example, I analyze the KWW relaxation function $\Psi\left(t\right)=\exp\left\{-\left(t/\tau\right)^{\beta}\right\}$ with $\beta<1$. It is apparent from equations (3) and (4) that the relaxation exponent function is given by:

$$\psi\left(t\right)=\left(\frac{t}{\tau}\right)^{\beta}, \tag{11}$$

and, when β and τ do not depend on time, the instantaneous relaxation time is given by:

$$\tau\left(t\right)=\frac{\tau^{\beta}}{\beta t^{\beta-1}}. \tag{12}$$

Therefore, when β and τ are constants, the instantaneous relaxation time is always an increasing function of time. Since $\mathrm{d}\tau\left(t_w\right)/\mathrm{d}t_w>0$, the KWW relaxation with constant β and τ belongs to Type I aging. The two-time instantaneous relaxation time for the KWW relaxation with constant β and τ is given by:

$$\tau\left(t',t_w\right)=\frac{\tau^{\beta}}{\beta\left(t_w+t'\right)^{\beta-1}}, \tag{13}$$

and therefore

$$\tau\left(0,t_w\right)=\frac{\tau^{\beta}}{\beta {t_w}^{\beta-1}}. \tag{14}$$

The type of aging thus can be identified by observing the protocol dependence of $\mathrm{d}\tau\left(0,t_w\right)/\mathrm{d}t_w$. For the Debye relaxation $\exp\left(-t/\tau\right)$ corresponding to the case $\beta=1$ of the KWW relaxation, the effective relaxation time is equal to τ and, therefore, if τ is a constant, the Debye relaxation shows no waiting time dependence of aging.

A power-law function $\Psi\left(t\right)=\left(\tau/t\right)^{\alpha}$ $(\alpha>0)$ is sometimes used to represent the behavior of relaxation [6, 8]. It is easy to confirm that if α and τ do not depend on time, the power-law relaxation belongs to Type I aging.

3. Free energy landscape and its relaxation

In many non-equilibrium systems such as super-cooled liquids, it is known that fast and slow modes of atomic dynamics are well separated, and that the free energy can be defined from the fast dynamics as a function of average atomic configuration. The dynamics of a representative point on the FEL is described by a stochastic Langevin equation with the FEL as the potential function [15]. One can use a reduced representation of the FEL, focusing only on the local minima of the FEL [15]. It should be emphasized that the essential part of the results in this paper does not depend on the representation. In the reduced representation, I denote by $\boldsymbol{P}\left(\boldsymbol{n},\boldsymbol{t}\right)$ the probability that the system is in basin $\boldsymbol{n}$ at time $\boldsymbol{t}$ and by $\boldsymbol{f}\left(\boldsymbol{n}\right)$ a physical quantity of basin $\boldsymbol{n}$. Then the observed value of the physical quantity is given by:

$$\mathcal{F}\left(\boldsymbol{t}\right)=\sum_{\boldsymbol{n}}\boldsymbol{f}\left(\boldsymbol{n}\right)\boldsymbol{P}\left(\boldsymbol{n},\boldsymbol{t}\right). \tag{15}$$

The time evolution of probability function $\boldsymbol{P}\left(\boldsymbol{n},\boldsymbol{t}\right)$ is assumed to be governed by a master equation:

$$\frac{\partial\boldsymbol{P}\left(\boldsymbol{n},\boldsymbol{t}\right)}{\partial\boldsymbol{t}}=\sum_{\boldsymbol{m}\neq\boldsymbol{n}}\boldsymbol{W}_{\boldsymbol{n},\boldsymbol{m}}\boldsymbol{P}\left(\boldsymbol{m},\boldsymbol{t}\right)-\ \boldsymbol{W}_{\boldsymbol{n},\boldsymbol{n}}\boldsymbol{P}\left(\boldsymbol{n},\boldsymbol{t}\right), \tag{16}$$

where $\boldsymbol{W}_{\boldsymbol{n},\boldsymbol{m}}$ is the transition rate from basin $\boldsymbol{m}$ to basin $\boldsymbol{n}$ and $\boldsymbol{W}_{\boldsymbol{n},\boldsymbol{n}}=\sum_{\boldsymbol{m}}\neq\boldsymbol{n}\boldsymbol{W}_{\boldsymbol{m}.\boldsymbol{n}}$. The dynamics is described by the transition rate matrix in equation (16), whose element can be typically written as:

$$\boldsymbol{W}_{\boldsymbol{n},\boldsymbol{m}}=\boldsymbol{w}_0\mathbf{exp}\left(-\frac{\Delta\boldsymbol{U}_{\boldsymbol{n},\boldsymbol{m}}\left(\boldsymbol{T}\right)}{\boldsymbol{k_B T}}\right), \tag{17}$$

where $\Delta\boldsymbol{U}_{\boldsymbol{n},\boldsymbol{m}}\left(\boldsymbol{T}\right)$ is the barrier height between two basins $\boldsymbol{m}$ and $\boldsymbol{n}$ and $\boldsymbol{w}_0$ is an attempt frequency determining the time scale of the walk. Note that $\boldsymbol{P}\left(\boldsymbol{n},\boldsymbol{t}\right)$ and hence $\mathcal{F}\left(\boldsymbol{t}\right)$ depend on temperature and that the total probability $\sum_{\boldsymbol{n}}\boldsymbol{P}\left(\boldsymbol{n},\boldsymbol{t}\right)$ is conserved.

The FEL depends on temperature by its definition, and it is natural to assume that the FEL will respond with delay to a temperature modulation. The temperature dependence of the FEL has been incorporated into the analysis by various concepts. Here, I exploit the simplest form that the jump rate (17) responds to the temperature change with delay:

$$\boldsymbol{W}\left(t\right)=\begin{cases} W_0 & (t<0) \\ W_1+\left(W_0-W_1\right)e^{-t/\tau_F} & (t\geqslant 0)\end{cases}, \tag{18}$$

where $W_0 \equiv w_0 \mathbf{exp}\left(-\frac{\Delta U_{n,m}(T_0)}{k_B T_0}\right)$ and $W_1 \equiv w_0 \mathbf{exp}\left(-\frac{\Delta U_{n,m}(T_1)}{k_B T_1}\right)$.

One can think of different models for the relaxation of the FEL like a delayed response of the barrier height or the fictive temperature [18–20]. It is easy to compare these models and confirm that the time dependence of the jump rate in these models are almost identical. Therefore, I exploit the simple model (18) for the description of the relaxation of the FEL in the following discussion. It should be emphasized that the essential results in the present paper do not depend on models for the delayed response of the FEL.

4. Aging of intermediate scattering function and its waiting time dependence

In order to investigate the aging of the intermediate scattering function, I consider a random walk model on a cubic lattice. I denote by $P(s,t)$ the probability that a random walker is at site s at time t when it started from the origin at time $t = 0$ and assume that it obeys the simple random walk master equation

$$\frac{\partial P(s,t)}{\partial t} = W(t)\left[\sum_d P(s+d,t) - zP(s,t)\right] \tag{19}$$

where d denotes a nearest neighbor site of site s and z is the coordination number of the lattice. Here, $W(t)$ is the jump rate of the random walker between two adjacent sites which reflects the barrier height produced by the FEL. For the temperature control equation (1), I assume that $W(t)$ relaxes as:

$$W(t) = W_1 + (W_0 - W_1)\, e^{-t/\tau_F}, \tag{20}$$

which satisfies $W(0) = W_0$ and $W(\infty) = W_1$. Equation (16) can be solved analytically by introducing a scaled time $\tilde{t}(t)$;

$$\tilde{t}(t) = \int_0^t W(t')\,\mathbf{d}t', \tag{21}$$

namely

$$d\tilde{t} = W(t)\,\mathbf{d}t. \tag{22}$$

Then, equation (19) reduces to:

$$\frac{\partial P(s,\tilde{t})}{\partial \tilde{t}} = \sum_d P(s+d,\tilde{t}) - zP(s,\tilde{t}). \tag{23}$$

I focus on the aging of the self-part of the intermediate scattering function $F_S(k,t)$ which is defined by:

$$F_S(k,t) = \sum_s e^{ik\cdot s} P(s,t), \tag{24}$$

where k is a wave vector. It is easy to show that $F_S(k,t)$ obeys:

$$\frac{\partial F_S(k,\tilde{t})}{\partial \tilde{t}} = -\gamma F_S(k,\tilde{t}), \tag{25}$$

where:

$$\gamma(k) = z - \sum_d e^{ik\cdot d}. \tag{26}$$

Therefore, $F_S(k,\tilde{t})$ is given by:

$$F_S(k,\tilde{t}) = F_S(k,0)\, e^{-\gamma(k)\tilde{t}} \tag{27}$$

and the two-time correlation function $\mathbf{C}(t',t_w) = F_S(k,t_w+t')/F_S(k,t_w)$ is written as:

$$\mathbf{C}(t',t_w) = \exp\left[-\gamma(k)\left\{\tilde{t}(t_w+t') - \tilde{t}(t_w)\right\}\right]. \tag{28}$$

Recalling the scaled time $\tilde{t}(t)$ defined by equation (21), I find:

$$\partial F_{\mathbf{S}}(\mathbf{k},\mathbf{t}) = F_S(k,0)\exp\left[-\gamma(k)\left\{\mathbf{W}_1 t + (\mathbf{W}_0 - \mathbf{W}_1) \times \tau_F\left(1 - e - t/\tau_F\right)\right\}\right] \tag{29}$$

and:

$$\mathbf{C}(t',t_w) = \exp\left[-\gamma(k)\left\{\mathbf{W}_1\mathbf{t}' + (\mathbf{W}_0 - \mathbf{W}_1) \times \tau_F e^{-t_w}/\tau_F\left(1 - e^{-t'/\tau_F}\right)\right\}\right]. \tag{30}$$

Figure 1 shows the t' and t_w dependence of $\mathbf{C}(t',t_w)$ for the temperature control: (a) T-down protocol and (b) T-up protocol. Apparently, the correlation function shows different waiting time dependence for T-up and T-down protocols.

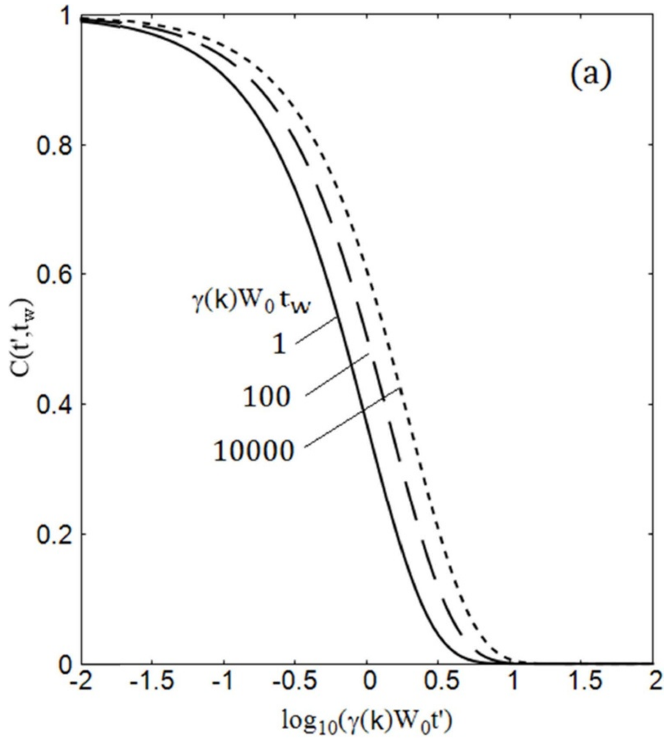
The instantaneous two-time relaxation time $\tau(t',t_w)$ is given by:

$$\tau(t',t_w) = -\left[\frac{\partial \ln \mathbf{C}(t',t_w)}{\partial t'}\right]^{-1}. \tag{31}$$

As shown in figure 2, $\tau(0,t_w)$ is an increasing function of t_w for T-down protocol and a decreasing function of t_w for T-up protocol.

Since the instantaneous two-time relaxation time $\tau(0,t_w)$ can be increasing or decreasing as t_w is increased, the aging is of Type II.

It should be mentioned that since the initial slope of $\tau(0,t_w)$ is determined by the relaxation time τ_F of the FEL, τ_F can be deduced from the t_w dependence of$\tau(0,t_w)$.



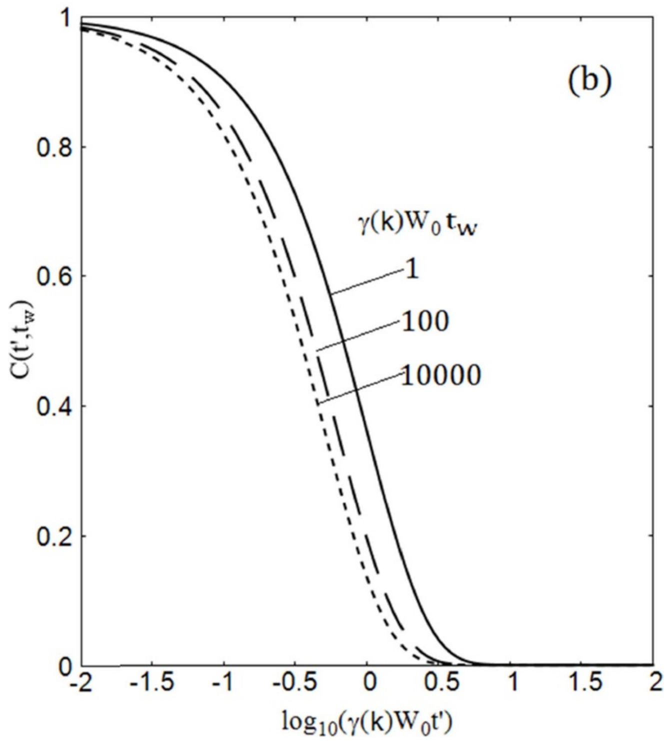


Figure 1. The t' and $t_w$dependence of $\mathbf{C}(t', t_w)$ for the temperature control: (a) T-down protocol ($\boldsymbol{W}_1/\boldsymbol{W}_0 = 0.5$) and (b) T-up protocol ($\boldsymbol{W}_1/\boldsymbol{W}_0 = 2.0$), where $\gamma(k)\,\boldsymbol{W}_0\tau_F = 100$.

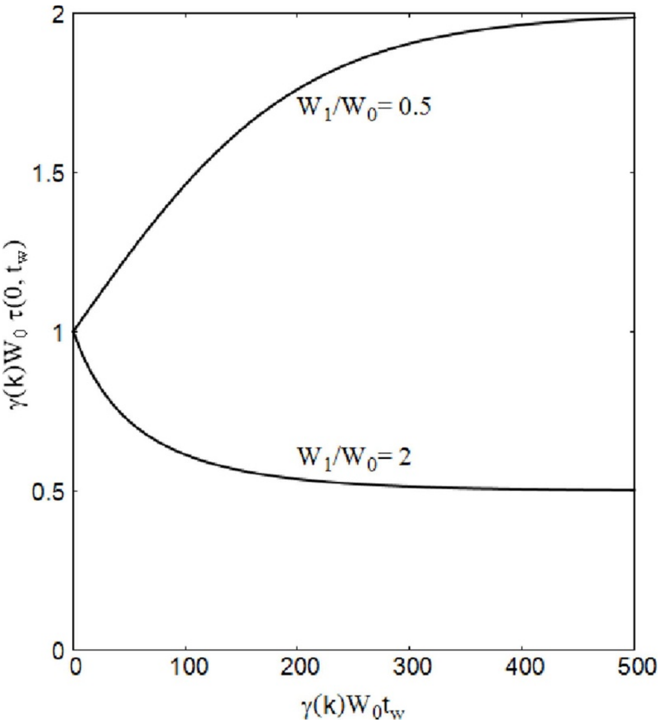


Figure 2. The t_w dependence of $\tau(0, t_w)$ for T-up protocol ($W_1/W_0 = 2.0$) and T-down protocol ($W_1/W_0 = 0.5$), where $\gamma(k) W_0 \tau_F = 100$.

5. Discussion

In this paper, I investigated the aging phenomenon on the basis of the free energy landscape theory of non-equilibrium systems and showed that (a) the aging can be classified into two types depending on the protocol dependence of the instantaneous relaxation time, (b) the relaxation of the FEL manifests itself as type II aging, (c) the instantaneous relaxation time can be either increasing or decreasing function of the waiting time and (d) the relaxation time of the FEL can be deduced from the waiting time dependence of the instantaneous relaxation time.

The most common relaxation function in non-equilibrium systems is the KWW function. The KWW relaxation with constant β and τ is said to represent the slow relaxation, in which the instantaneous relaxation time is always an increasing function of the waiting time. Therefore, in order to explain the observation in which the instantaneous relaxation time for T-up protocol is an increasing function of the waiting time, a waiting time dependence of exponent β has been considered [10]. The waiting time dependence of β and τ of the KWW function may be understood by the relaxation of the FEL. It is clear that the opposite dependence of the instantaneous relaxation time on the waiting time for T-up and T-down protocols can be explained by the delayed response of the FEL.

The present result indicates that the slow relaxation may well be fitted by:

$$\Phi(t) = \Phi(0)\, e^{-\tilde{t}(t)} \tag{32}$$

with $\tilde{t}(t) = A\left[W_1 t + (W_0 - W_1)\tau_F\left(1 - e^{-t/\tau_F}\right)\right]$, where A is a positive constant. In fact, it is straightforward to confirm that equation (32) behaves almost identical to the KWW relaxation function.

It should be emphasized that the present formalism is very much robust and can be applied to any other aging phenomena, including the dielectric relaxation, the dielectric response [14, 21, 22] and the order parameter dynamics for the Landau model [23].

Data availability statements

All data that support the findings of this study are included within the article.

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ORCID iD

Takashi Odagaki https://orcid.org/0000-0002-9991-128X

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