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**Key Points:**
- Anomalous high unblocking temperature of viscous remanence is due to slow relaxation
- Stretched exponential function explains published data on magnetic relaxation
- A nonlinear time-temperature relationship is derived

**Supporting Information:**
- Supporting Information S1
- Table S1

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# Stretched exponential relaxation of viscous remanence and magnetic dating of erratic boulders

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**Abstract** Viscous remanence continuously increases with the duration of reorientation of rocks, and the remanence gets partially overprinted in rocks parallel to the Earth's magnetic field. This overprinted viscous remanence is unblocked at a certain temperature that enables the estimation of the time required for the rock to acquire the magnetism, by assuming the exponential law of Néel's single-domain theory. However, previous results of dating the rocks by the exponential law have shown older ages than radiometric or cosmogenic exposure ages. Néel's exponential decay law is applicable to a system whose magnetic grains have an identical relaxation time. However, in real systems, the expected behavior is not usually observed because relaxation times vary for individual grains. Moreover, the variation of viscous remanence with the logarithmic law for a distribution of relaxation times is predicted to be concave downward. Here we found that the stretched exponential law, $\exp\{-(t/\tau)^{1-n}\}$ with $0 \le n < 1$, explains previously published data on viscous decay. The observed stretched exponential relaxation can be interpreted in terms of the global relaxation of a system containing many relaxing species, each of which decays exponentially in time with a specific fixed relaxation rate. Using this law, we derived an extended time-temperature relationship of magnetite involving the Néel's exponential decay law with $n = 0$ and a system containing many relaxing times with variable $n$. The extended time-temperature relationship shows that the age of a coral tsunami boulder with high anomalous unblocking temperatures can be fitted with an assigned radiometric age.

## 1. Introduction

Ages of erratic boulders reworked by geological and archeological events (e.g., cliff slump, moraine deposition, and tsunami events) contribute to hazard awareness and ancient history. In rock magnetism and paleomagnetism, the reworked age of erratic boulders is usually determined considering an unblocking temperature of secondary viscous remanence [*Borradaile*, 1996, 1998; *Crider et al*., 2015; *Kent*, 1985; *Middleton and Schmidt*, 1982; *Muxworthy et al*., 2015; *Sato et al*., 2014; *Tyson-Smith and Verosub*, 1994]. Most rocks contain an aggregate of ferrimagnetic minerals that record the geomagnetic field as natural remanence. If such rocks are emplaced or reworked, a secondary viscous remanence overprints the primary characteristic remanence and the viscous remanence increases in the present Earth's magnetic field. A conventional approach for determining the age by viscous remanence is to establish a time-temperature relationship (Pullaiah's nomogram) by assuming Néel's relaxation theory of single-domain magnetite [*Néel*, 1949, 1955; *Pullaiah et al*., 1975]. Relaxation time is a measure of the time taken by a grain to overcome its energy barrier by thermal activations. If single-domain magnetite can acquire viscous remanence in a magnetic field at low temperature ($T_A$) over a long relaxation time ($t_A$), the remanence is demagnetized in the absence of the magnetic field at high temperature ($T_D$) over a short relaxation time ($t_D$) and is given by

$$\frac{T_D \ln(Ct_D)}{J_S^2(T_D)} = \frac{T_A \ln(Ct_A)}{J_S^2(T_A)}, \tag{1}$$

where $C$ is a frequency factor usually taken as $10^{10}$ Hz and $J_s(T)$ is the saturation magnetization. The temperature $T_D$ at which the viscous remanence component is completely erased is termed unblocking temperature. In equation (1), the coercivity of magnetite is dominated by shape anisotropy and is proportional to $J_S(T)$ [*Pullaiah et al*., 1975]. Variations in $J_s(T)$ are determined by $J_s(T) = J_s(T_{293K})\{(T_c - T)/(T_c - T_{293K})\}^{0.43}$, where $T_c$ is the Curie temperature for magnetite in kelvin, $J_s(T_{293K}) = 4.8 \times 10^5$ A/m [*Dunlop and Özdemir*, 1997]. The reworked age can be determined through experimental time-temperature combinations for removing viscous remanence at ambient temperature [e.g., *Borradaile*, 1996]. *Sato et al.* [2014] used viscous

remanence dating to estimate reworked ages of two coral boulder moved by tsunamis. The timing of tsunamis estimated from unblocking temperatures of viscous remanence was compared with $^{14}C$ accelerator mass spectrometry ages obtained from the surface of well-preserved coral boulders [*Araoka et al.*, 2013]. *Crider et al.* [2015] analyzed the viscous remanence of granitic boulders in glacial moraines. Although boulders might be reworked by degradation of older moraines, $^{36}Cl$ cosmogenic exposure age of the top surface of the largest moraine boulders [*Porter and Swanson*, 2008] was used as a comparable measure of viscous remanence age. However, the viscous remanence of boulders having higher unblocking temperature than the one calculated from Pullaiah's nomogram is problematic; thus, viscous remanence yielded older ages than the $^{14}C$ or $^{36}Cl$ cosmogenic ages. Such discrepancies have been reported in previous studies involving calibration by many techniques (e.g., historical records, erosion rate, conodont alteration index, and paleosecular variation) [*Borradaile*, 1996, 1998; *Kent*, 1985; *Muxworthy et al.*, 2015]

Following Néel's theory, time-dependent viscous remanence of a sample containing a large number of identical single-domain grains changes exponentially. In general, a natural rock contains several ferrimagnetic grains with widely varying sizes and shapes as described by their grain size distributions. In such an aggregate, acquisition and decay of viscous remanence is regarded as the result of the superposition of different relaxation times [*Saito and Murayama*, 1987]. The summation of the lognormal distribution of exponential relaxation times for single-domain grains leads to the logarithmic behavior: $S \ln(t)$, where $S$ is the temperature dependence coefficient and $t$ is elapsed time [*Dunlop*, 1983; *Street and Woolley*, 1949]. However, experimental data of magnetic relaxations have shown concave downward or upward slopes of logarithmic approximations [*Dunlop*, 1983; *Halgedahl*, 1993; *Kelso and Banerjee*, 1994; *Moskowitz*, 1985; *Smith*, 1984; *Tivey and Johnson*, 1984; *Trukhin et al.*, 2003; *Williams and Muxworthy*, 2006]. Moreover, although *Walton* [1980] has established an alternative time-temperature relationship based on a lognormal grain size distribution of magnetite, the equation is not reducible to Pullaiah's nomogram for single-domain grains. The presence of coarse magnetic grains (pseudo single-domain and multi-domain) in rocks is key to solving the discrepancy between the relaxation theory and experimental results. It has been believed that nonexponential or nonlogarithmic relaxation is caused by the presence of coarse grains where large domains with defects relax slowly [*Chamberlin and Scheinfein*, 1993], yielding anomalous high unblocking temperatures. Therefore, an alternative relaxation theory is needed for understanding the complex magnetic aggregation.

In physics, relaxation is believed to be an aging process in which a system gradually changes from a far-from-equilibrium to an equilibrium state [e.g., *Chamberlin and Scheinfein*, 1993]. It is well known that structural relaxation of glasses, spin glasses, and other complex system states generally exhibits nonexponential and nonlogarithmic relaxation, being often described in terms of the stretched exponential law [e.g., *Kohlrausch*, 1854; *Ngai*, 1998]. *Chamberlin et al.* [1984] found that the decay of thermoremanence within the spin glass region exhibits a stretched exponential law rather than other relaxation laws. The features of spin glasses, such as remanence, hysteresis, and time-dependent effects, have been observed in rock magnetism as described by Néel's theory [*Ford*, 1982; *Wohlfarth*, 1977]. In this study, we revisit Néel's theory to fit previously reported relaxation data by a stretched exponential law. Using this law, we derive an extended time-temperature relationship of magnetite, which could reproduce Néel's exponential model and explain discrepancies.

## 2. Theory

Classically, relaxation processes are described by the exponential function:

$$\phi(t) = \exp\left(-\frac{t}{\tau}\right), \tag{2}$$

which is referred to as the Maxwell-Debye relaxation or Néel's relaxation [*Maxwell*, 1867; *Néel*, 1949]. In equation (2), the relaxation function $\phi(t)$ possesses the characteristic relaxation rate $\tau$ and the normalized initial condition $\phi(0) = 1$. The exponential function in equation (2) fulfills a first-order linear differential equation:

$$\frac{d\phi(t)}{dt} = -\phi(t)/\tau. \tag{3}$$

As relaxation rate is constant, decay rate is independent of time. However, dynamical systems showed pronounced deviations from the ideal exponential pattern. As early as 1863, *Kohlrausch* [1863] described mechanical creep of glassy fibers by the following stretched exponential function:

$$\phi(t) = \exp\left\{-\left(\frac{t}{\tau}\right)^{1-n}\right\}, 0 \le n < 1, \tag{4}$$

where $n$ is a stretching exponent. *Williams and Watts* [1970] described dielectric relaxation in polymers using equation (4). The "Brinkman Report" [*Brinkman*, 1986] stated that "there seems to be a universal function that slow relaxations obey. If the system fluctuates out of equilibrium, it returns according to the function $\exp\{-(t/\tau)^{1-n}\}$. Unfortunately, this is not a mathematical expression that is frequently encountered in physics, so little idea exists of what the underlying mechanisms are." Since then, a large number of relaxation systems (ranging from earthquakes, galactic light emissions, and biological excitation to economics and scientific citations) have been found to exhibit this type of function [*Laherrère and Sornette*, 1998], and numerous underlying physical mechanisms have been proposed for their derivations [*Phillips*, 1996]. *Phillips* [1996] investigated an excited singlet electron in a fluorophore molecule and reported that the presence of progressively depleted random sinks that capture electron excitations can modify a spontaneous decay process represented by equation (2), such that the relaxation rate is dependent on time, stretching the decay:

$$\frac{\mathrm{d}\phi(t)}{\mathrm{d}t} = -\phi(t)\cdot\psi t^{-n}, \tag{5}$$

where $\psi$ is a characteristic constant. Now we consider relaxation function $\phi(t)$ as the relaxation of remanence $J(t)$, then $\mathrm{d}J(t)/\mathrm{d}t = -W(t)J(t)$, where $W(t)$ is the time-dependent relaxation rate. The relaxation rate can be described by the power law

$$W(t) = \psi t^{-n}. \tag{6}$$

In 1979, *Funabashi and Hamill* [1979] proposed a detailed derivation of the time-dependent relaxation rate, equation (6), with the continuous time random walk model developed for the anomalous electron transit time dispersion in amorphous solids [*Scher and Montroll*, 1975]. Since then, this type of relaxation rate has been assumed in a wide range of relaxation studies [*Shlesinger and Hughes*, 1981; *Hara and Okayama*, 1988; *Ngai and Strom*, 1988; *Colmenero et al.*, 1993; *Ulrich et al.*, 2003; *Porto*, 2005; *Kawada et al.*, 2006]. Using the power law relaxation rate $W(t)$, we can obtain the relaxation of remanence:

$$J(t)/J_0 = \exp\left\{-\psi t^{1-n}/(1-n)\right\}. \tag{7}$$

Equation (7) can be rewritten as

$$J(t) = J_0 \exp\left\{-\left(\frac{t}{\tau}\right)^{1-n}\right\}, 0 \le n < 1, \tag{8}$$

where $\psi = (1-n)/\tau^{1-n}$ [*Berberan-Santos et al.*, 2005; *Berberan-Santos*, 2008]. For $n = 0$, equation (8) is reduced to a simple Néel's exponential relaxation [*Ngai and Strom*, 1988; *Ulrich et al.*, 2003]. The dynamics of slow relaxation is expressed by a longer relaxation time instead of simple relaxation time [*Ngai*, 1998; *Hara and Tamura*, 2013].

In the field of rock magnetism and paleomagnetism, estimation of the age of viscous remanence may be complicated if rocks require anomalously high unblocking temperatures or long relaxation times to erase remanence, thus leading to overestimation of the age. The presence of high unblocking temperature and long relaxation time indicates that the rate of observed magnetic relaxation is slower than that from the conventional Néel's relaxation theory. Therefore, such a slow relaxation of the aggregate of magnetic particles should be represented by the stretched exponential law. The aggregate of magnetic particles may yield a continuous distribution of relaxation times, and then the stretched exponential function (8) can be mathematically expressed as a superposition of exponential terms as follows:

$$J(t) = \int_0^\infty \exp\left(-\frac{t}{\tau}\right)\rho(\tau)\mathrm{d}\tau, \tag{9}$$

where $\rho(\tau)$ is a continuous distribution of relaxation times. Such continuous distributions of relaxation time due to varying grain sizes have been observed in natural rock samples as well as in artificial products.

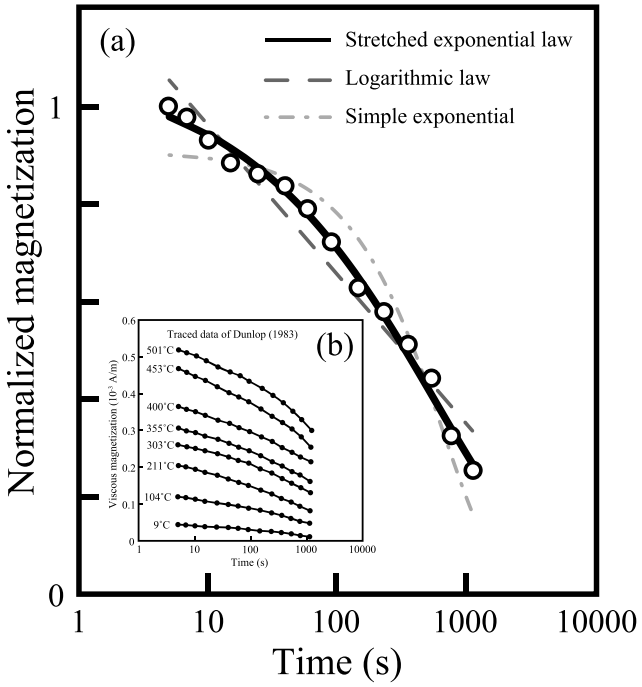


**Figure 1.** (a) Magnetic decay at 9°C for synthetic fine magnetite (data from *Dunlop* [1983]). It has been confirmed that the stretched exponential law (solid line) is a more reliable fit to the observations (open circles) than the logarithmic (dashed line) and simple exponential laws (chain line). (b) Traced lines show concave downward slopes [*Dunlop*, 1983].

Therefore, the mathematical equivalent of equations (8) and (9) predicts a stretched exponential relaxation of remanence in geological materials. *Plonka* [1983] presented a detailed derivation of the distribution through Laplace transformation. The decay of remanence should follow equation (8), with $n = 0$ for identical single-domain particles and $0 < n < 1$ for aggregates with a continuous distribution of relaxation time in rocks [*Porto*, 2005; *Ulrich et al.*, 2003]. For $n \geq 1$, the relaxation law is described by the power law, which is independent of size distribution [*Porto*, 2005; *Ulrich et al.*, 2003]. The relaxation time, which depends on the temperature $T$ of an aggregate, is scaled by stretching exponent $n$ and given by the following simple form:

$$\tau^* = \tau^{1-n} = \left\{ \frac{1}{C} \exp\left( \frac{v h_c J_s}{kT} \right) \right\}^{1-n}, \quad (10)$$

where $v h_c J_s / 2 = E$ is the height of the energy barrier prohibiting the rotation of electron spin moments, $h_c$ is coercivity, $k$ is the Boltzmann's constant, and the scaled relaxation time $\tau^*$ replaces the longer relaxation time $\tau$. If a system features different relaxation times (e.g., an aggregate of single-domain, pseudo single-domain, and multi-domain grains), the relaxation time can be described by equation (10). The stretching exponent $n$ in equation (10) is a dispersion factor from an aggregate of ideal single-domain grains, where the energy barrier and frequency factor is scaled as $(1 - n) \times E$ and $C^{1-n}$ [*Benatar et al.*, 1993; *Chen*, 2003]. A large energy barrier $E$ is expected to increase $\tau$ of the aggregate [*Chamberlin and Scheinfein*, 1993]. Moreover, the frequency factor $C$ has been found to be strongly dependent on the morphology of individual grains, such that small differences in size or shape can lead to variations in the relaxation rates [*Berndt et al.*, 2015; *Krause et al.*, 2009]. At a given fixed $T$, $\tau$ ($T$, $E$, $C$) depends on $E$ and $C$ [*Prester et al.*, 2011]. Thus, $n$ is characterized by both factors. In equation (10), $\tau^*$ scaled by $1 - n$ is the real relaxation time for the aggregate. If an aggregate of magnetic grains with a particular ($v$, $h_c$) has a relaxation time $t_A^*$ at temperature $T_A$, then its relaxation time $t_D^*$ at $T_D$ is given by

$$\frac{T_D \ln\left\{ C \left( t_D^* \right)^{\frac{1}{1-n}} \right\}}{J_S^2(T_D)} = \frac{T_A \ln\left\{ C \left( t_A^* \right)^{\frac{1}{1-n}} \right\}}{J_S^2(T_A)}. \quad (11)$$

If $n$ is determined, the unknown $t_A^*$ is estimated to be the true age at $T_A$ when $T_D$ is known (from thermal demagnetization experiments) with a duration time of $t_D^*$.

## 3. Results

We reanalyzed previously published data on viscous remanence decay regarding the relaxation of magnetite following the stretched exponential law in equation (8). Figure 1a shows a plot of typical decay of 76 nm synthetic magnetite in the single-domain range against logarithmic time at 9°C (data traced form *Dunlop* [1983]). The slope of relaxation was concave downward of the logarithmic approximation (Figure 1b). Instead of the slope of the logarithmic and simple exponential laws, the relaxation function satisfies a stretched exponential form throughout the time range. The best fit value of stretching exponent $n = 0.54$ was calculated using IGOR

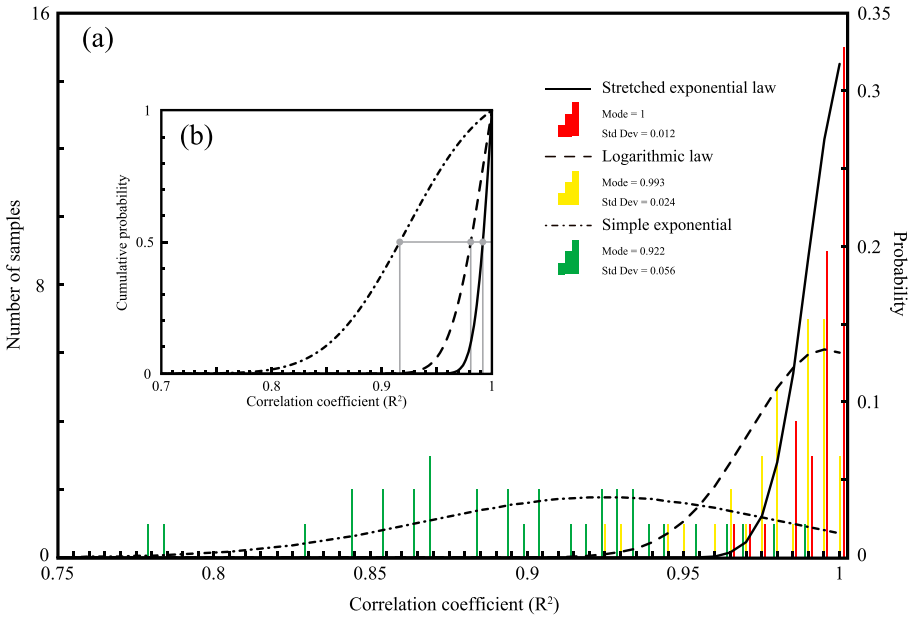


**Figure 2.** (a) Histograms of correlation coefficients (color bars) and their probability distribution curves (solid, dashed, and chain lines). (b) Cumulative probability plot for three different laws. Gray points and lines show half width at half maximums of each plot.

Pro 6.33 J (WaveMetrics, Inc., USA). Color bars in Figure 2a show the histogram of correlation coefficients ($R^2$) obtained directly by fitting curves of viscous remanence decay from previously published data for synthetic and natural rock samples ($N = 34$). The red bars represent correlation coefficients of stretched exponentials, while the green and yellow bars represent logarithmic and simple exponentials (Table S1 in the supporting information; data from *Dunlop* [1983], *Halgedahl* [1993], *Kelso and Banerjee* [1994], *Tivey and Johnson* [1984], *Trukhin et al.* [2003], and *Williams and Muxworthy* [2006]). In the range of 0.962 to 1, correlation coefficients of stretched exponential laws are distributed with a mode of 1. The correlation coefficients of the logarithmic law range between 0.922 and 0.997 with a mode of 0.993 and that for the simple exponential exhibits scattered distribution between 0.776 and 0.986 with a mode of 0.922. The most reasonable fits were obtained by stretched exponentials for 31/34 (91.2%) samples. We also obtained probability distribution curves by fitting truncated normal distribution to each histogram. The curve of stretched exponential law exhibits a sharp central peak, while the curves of logarithmic law and simple exponential are broad. In Figure 2b, we show the cumulative probability curves of the three laws. The half width at half maximum of the stretched exponential law is 0.009, which is smaller than those of the logarithmic and simple exponential laws at 0.019 and 0.084, respectively. It is concluded that the stretched exponential law with the value of $n$ is the most effective for explaining observations.

The applicability of the extended time-temperature relationship of magnetite in equation (11) was tested using a 200 metric ton coral tsunami boulder from Ishigaki Island. The reworked age of the boulder was determined to be $2.69 \pm 0.02$ kyr B.P. by $^{14}$C dating. Remanence carriers in the boulder were single-domain and/or pseudo single-domain magnetite [*Sato et al.*, 2014]. In the dating of viscous remanence, low-temperature demagnetization was applied as pretreatment in order to demagnetize defectless multi-domain sized magnetite particles, isolating the single-domain-like remanence [e.g., *Ozima et al.*, 1964; *Dunlop and Argyle*, 1991]. Although the remanence of multi-domain and pseudo single-domain grains of unpinned or loosely pinned domain walls can be removed through the low-temperature demagnetization, the remanence strongly pinned domain walls are excluded [*Özdemir and Dunlop*, 1999]. The pinning may explain the remanence observed in multi-domain and single-domain-like remanence in pseudo single-domain grains [e.g., *Kubo et al.*, 2015]. Our estimated unblocking temperature of 145°C translates to an age of 14 kyr B.P. in Pullaiah's nomogram, which disagrees with the $^{14}$C age [*Sato et al.*, 2014]. In our experiments, stepwise thermal demagnetization with a 5°C increment was used for the demagnetization temperature range of 100–160°C with a duration of 300 s. For the 80–100°C range, a 10°C interval was used and other points were 200°C, 250°C,

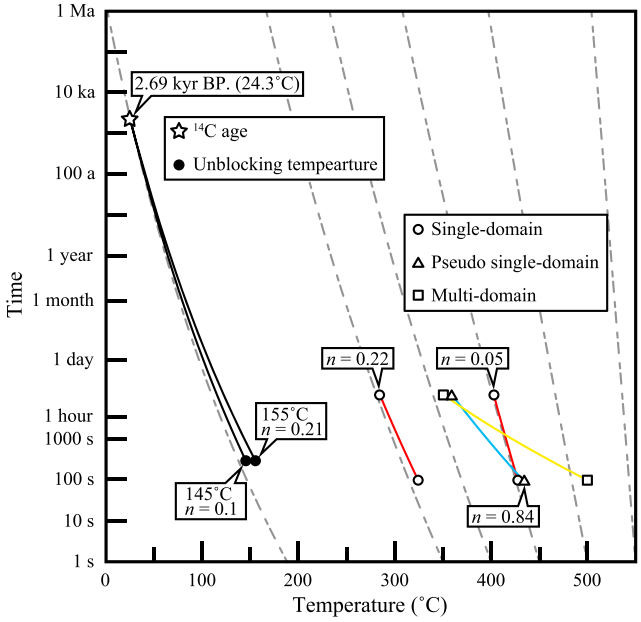


**Figure 3.** A comparison of corresponding (*t*, *T*) values. Black circles show demagnetization temperatures (unblocking temperatures) of viscous remanence of a coral tsunami boulder with a duration of 300 s, and contours are joined using extended *t-T* relation with its radiocarbon age (white star) at average annual temperature of Ishigaki Island, Japan (24.3°C). The point of 155°C is obtained from our experiment, and the data of 145°C are given by *Sato et al.* [2014]. The single-domain data pairs (white open circles with red lines) parallel to the contour of Pullaiah's nomogram for magnetite (gray dashed lines) and two stretched exponents *n* have small values. The pseudo single-domain and multi-domain data pairs (triangles with a blue line and squares with a yellow line) show less parallel contours. These data were obtained from *Dunlop and Özdemir* [2000].

300°C, and 400°C. Figure S1 shows an example of the resulting changes in vector components. The viscous remanence component is parallel to the present Earth's magnetic field up to its unblocking temperature of 155°C. The original component as a characteristic remanence direction is then isolated from the maximum temperature of 400°C. In Pullaiah's nomogram, an unblocking temperature of 155°C corresponds to an age of 13 kyr B.P. for the average temperature of 24.3°C at Ishigaki Island. As shown in the time-temperature nomogram in Figure 3, both unblocking temperatures of 145°C and 155°C are higher than the expected unblocking temperature in the contours of Pullaiah's nomogram (dashed line). In order to calibrate two higher unblocking temperatures, we applied the extended time-temperature relation. The calibrated results showed good agreement with the radiocarbon age when $n = 0.1$ and 0.21 (Figure 3).

## 4. Discussion

It is commonly observed that remanence does not change exponentially with time because the relaxation behavior is the summation of various relaxation times in any real sample [*Saito and Murayama*, 1987]. The acquisition or decay of viscous remanence in proportion to the logarithm of elapsed time could not explain the concave upward or downward slopes of magnetic relaxation. Moreover, the logarithmic law predicts that the intensity of magnetization of the sample eventually reaches an infinite value [*Street and Woolley*, 1949]. In order to establish the fundamental base for magnetic relaxation, we applied the stretched exponential law to the data. In complex systems, such as aggregates of single-domain, pseudo single-domain, and multi-domain grains, the relaxation behavior can be explained by stretched exponential law with a transformation from $\tau$ to $\tau^*$ [*Hara and Tamura*, 2013]. The relaxation rate of the power law describing the stretched exponential law has been shown to reflect the distribution of relaxation time in complex systems [*Plonka*, 1983, 1986]. The power law has been explained to result from the continuous time random walk model developed for dispersion in amorphous solids and utilized in describing the decay of excess electrons trapped in rigid glassy matrices [*Hamill and Funabashi*, 1977; *Funabashi and Hamill*, 1979]. In studies of rock magnetism, even though the low-temperature demagnetization can minimize the nonliner relaxation from coarse grains (pseudo single-domain and multi-domain) in a natural rock sample, previous researches have shown anomalously high unblocking temperatures [e.g., *Sato et al.*, 2014]. The high unblocking temperature persists even after repeated low-temperature demagnetization, being consistent with dislocation pinning of domain walls in non-single-domain grains [*Crider et al.*, 2015]. From the domain wall pinning model, an interest in the Barkhausen effect has grown considerably in connection with complex systems. The Barkhausen effect is basically governed by a moving domain wall [*Bertotti and Montorsi*, 1990], where the distributions of domain wall velocity, Barkhausen jump amplitudes, and their duration are well fitted by power laws [*Durin and Zapperi*, 2000; *Zapperi et al.*, 1998]. When domain walls pinned at a specific temperature in a magnetic field move toward a demagnetized state on heating in zero field to an unblocking temperature, Barkhausen jumps yield high unblocking temperatures

for complete demagnetization [*Dunlop*, 1983]. The presence of random defects pinning the movement of magnetic domains can modify a spontaneous relaxation process of remanence, where the stretching exponent is fully determined from the variation in energy barrier and Arrhenius-Néel prefactor characterizing the structure of the system [*Berndt et al*., 2015; *Chamberlin and Scheinfein*, 1993; *Colmenero et al*., 1993; *Hara and Okayama*, 1988; *Kawada et al*., 2006; *Krause et al*., 2009; *Ngai and Strom*, 1988; *Porto*, 2005; *Prester et al*., 2011; *Shlesinger and Hughes*, 1981; *Ulrich et al*., 2003]. It has been suggested that the pseudo single-domain behavior, exhibiting a blended response of single-domain and multi-domain particles [*Dunlop*, 2012; *Dunlop and Özdemir*, 2000], might arise from the Barkhausen jumps. The previous magnetic relaxation theory could not explain the relaxation behavior of natural samples without the stretching exponent. In our results, the stretched exponential law is the most suitable for describing the experimental decay of viscous remanence (Figure 2). Although previous researchers have used synthetic magnetite in order to isolate the effect of grain distribution [e.g., *Dunlop*, 1983], it is difficult to synthesize ideal magnetite particles with well-controlled grain sizes and shapes [*Yu et al*., 2002]. Overall, the magnetic relaxation behavior of synthetic and natural magnetite should be interpreted by grain distribution.

From the stretched exponential law, we derived the extended time-temperature relationship in equation (11). Natural rocks may yield older magnetic relaxation ages due to the anomalously high unblocking temperatures for magnetic overprints [*Borradaile*, 1996, 1998; *Kent*, 1985; *Middleton and Schmidt*, 1982; *Sato et al*., 2014; *Walton*, 1980]. The Pullaiah's relation based on the Arrhenius-Néel law is applicable only to systems with identical single-domain magnetite grains but not to aggregates containing larger grains with multiple magnetic domains [*Dunlop*, 2012; *Dunlop and Özdemir*, 1993, 2000; *Walton*, 1980]. The extended relationship could fit the unblocking temperature of viscous remanence with a varied $n$ value ($0 \leq n < 1$), which is a convenient measure of magnetic relaxation [*Chamberlin et al*., 1984; *Colmenero et al*., 1993; *Ngai*, 2007]. The relaxation time $\tau^*$ scaled by $n$ reflects the complex structure of the magnetic aggregate. In addition, our extended time-temperature relationship in equation (11) could adapt the single-domain grains with $n = 0$, although an alternative relation proposed by *Walton* [1980] could not explain Pullaiah's nomogram.

Figure 3 shows the extended relation for a coral tsunami boulder. Experimental results showed a better agreement with the nomogram based on the stretched exponential law. The reworked ages of boulders determined by magnetic relaxation could possibly be explained by the stretched exponential law. In order to evaluate the dependence of the time-temperature relationship on grain size, we investigated single-domain, pseudo single-domain, and multi-domain contours of magnetite (data from *Dunlop and Özdemir* [2000]). For single-domain grains of 40 nm, lines joining pairs of points on a graph were nearly parallel to contours of Pullaiah's nomogram with $n = 0.05$ and 0.22. This result suggests that the time-temperature relationship is affected by small differences in grain size because the synthetic single-domain magnetite could not be completely fitted to the contours of Pullaiah's nomogram. For pseudo single-domain grains of 20 μm and multi-domain grains of 135 μm, the lines joining blocking and unblocking temperature pairs become increasingly oblique to the contours of Pullaiah's nomogram, and these unblocking temperatures were anomalously high compared to identical single-domain values. When we fit the extended time-temperature contours to pseudo single-domain grains, exhibiting a blended response of single-domain and multi-domain grains [*Dunlop*, 2012; *Dunlop and Özdemir*, 2000], the value of stretching exponential $n$ was 0.84. For the multi-domain sample, $n$ did not have a solution in the range of $0 \leqq n < 1$. Therefore, the stretched exponential law is interpreted to explain the relaxation of an aggregate containing single-domain or single-domain-like responses.

## 5. Conclusions

In paleomagnetism and rock magnetism, the Pullaiah's time-temperature relationship for single-domain magnetites has been used to predict reworked ages using viscous remanence of erratic boulders. However, estimated ages have been shown to be older than ages from absolute or relative dating. To overcome this problem, we revisited Néel's theory and found that magnetic relaxation behavior is well described by the stretched exponential function, which takes into account the distributions of relaxation constants or grain sizes. The stretched exponential law provides an extended time-temperature relationship to explain the anomalous overestimation of ages caused by the presence of pseudo single-domain and/or multi-domain

grains, and our extended relation could calibrate the age of a boulder moved by a paleotsunami. With further insights into the mechanisms of older age, the dating of viscous remanence can be developed into a more reliable and quantitative dating method for revealing the absolute ages of materials.

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## References

Araoka, D., Y. Yokoyama, A. Suzuki, K. Goto, K. Miyagi, K. Miyazawa, H. Matsuzaki, and H. Kawahata (2013), Tsunami recurrence revealed by *Porites* coral boulders in the southern Ryukyu Islands, Japan, *Geology*, *41*, 919–922, doi:10.1130/G34415.1.

Benatar, L. E., D. Redfield, and R. H. Bube (1993), Interpretation of the activation energy derived from a stretched-exponential description of defect density kinetics in hydrogenated amorphous silicon, *J. Appl. Phys.*, *73*, 8659, doi:10.1063/1.353378.

Berberan-Santos, M. N. (2008), A luminescence decay function encompassing the stretched exponential and the compressed hyperbola, *Chem. Phys. Lett.*, *460*, 146–150, doi:10.1016/j.cplett.2008.06.023.

Berberan-Santos, M. N., E. N. Bodunov, and B. Valeur (2005), Mathematical functions for the analysis of luminescence decays with underlying distributions 1. Kohlrausch decay function (stretched exponential), *Chem. Phys.*, *315*, 171–182, doi:10.1016/j.chemphys.2005.04.006.

Berndt, T., A. R. Muxworthy, and G. A. Paterson (2015), Determining the magnetic attempt time $\tau_0$, its temperature dependence, and the grain size distribution from magnetic viscosity measurements, *J. Geophys. Res. Solid Earth*, *120*, 7322–7336, doi:10.1002/2015JB012283.

Bertotti, G., and A. Montorsi (1990), Dependence of Barkhausen noise on grain size in ferromagnetic materials, *J. Magn. Magn. Mater.*, *83*, 214–216, doi:10.1016/0304-8853(90)90490-H.

Borradaile, G. J. (1996), An 1800-year archeological experiment in remagnetization, *Geophys. Res. Lett.*, *23*, 1585–1588, doi:10.1029/96GL01575.

Borradaile, G. J. (1998), Rock magnetic constraints on long-term cliff-slump rates and coastal erosion, *Géotechnique*, *48*, 271–279, doi:10.1680/geot.1998.48.2.271.

Brinkman, W. F. (1986), *Physics Through the 1990s Condensed-Matter Physics*, pp. 328, National Academy of Science, Washington D. C.

Chamberlin, R. V., and M. R. Scheinfein (1993), Slow magnetic relaxation in iron: A ferromagnetic liquid, *Science*, *260*, 1098–1101, doi:10.1126/science.260.5111.1098.

Chamberlin, R. V., G. Mozurkewich, and R. Orbach (1984), Time decay of the remanent magnetization in spin-glasses, *Phys. Rev. Lett.*, *52*, 867, doi:10.1103/PhysRevLett.52.867.

Chen, R. (2003), Apparent stretched-exponential luminescence decay in crystalline solids, *J. Lumin.*, *102–103*, 510–518, doi:10.1016/S0022-2313(02)00601-4.

Colmenero, J., A. Alegría, I. Tellería, and K. L. Ngai (1993), Extended Kronmüller model for cooperative relaxations in metallic glasses, *Phys. Rev. B*, *47*, 5041, doi:10.1103/PhysRevB.47.5041.

Crider, J. G., D. M. Globokar, R. F. Burmester, and B. A. Housen (2015), Unblocking temperatures of viscous remanent magnetism in displaced granitic boulders, Icicle Creek glacial moraines (Washington, USA), *Geophys. Res. Lett.*, *42*, 10,647–10,654, doi:10.1002/2015GL066381.

Dunlop, D. J. (1983), Viscous magnetization of 0.04–100 μm magnetites, *Geophys. J. R. Astron. Soc.*, *74*, 667–687, doi:10.1111/j.1365-246X.1983.tb01899.x.

Dunlop, D. J. (2012), Magnetic recording in rocks, *Phys. Today*, *65*, 31–37, doi:10.1063/PT.3.1604.

Dunlop, D. J., and K. S. Argyle (1991), Separating multidomain and single-domain-like remanences in pseudo-single-domain magnetites (215–540 nm) by low-temperature demagnetization, *J. Geophys. Res.*, *96*, 2007–2017, doi:10.1029/90JB02338.

Dunlop, D. J., and Ö. Özdemir (1993), Thermal demagnetization of VRM and pTRM of single domain magnetite: No evidence for anomalously high unblocking temperatures, *Geophys. Res. Lett.*, *20*, 1939–1942, doi:10.1029/93GL02137.

Dunlop, D. J., and Ö. Özdemir (1997), *Rock Magnetism: Fundamentals and Frontiers*, pp. 573, Cambridge Univ. Press, Cambridge and New York.

Dunlop, D. J., and Ö. Özdemir (2000), Effect of grain size and domain state on thermal demagnetization tails, *Geophys. Res. Lett.*, *27*, 1311–1314, doi:10.1029/1999GL008461.

Durin, G., and S. Zapperi (2000), Scaling exponents for Barkhausen avalanches in polycrystalline and amorphous ferromagnets, *Phys. Rev. Lett.*, *84*, 4705, doi:10.1103/PhysRevLett.84.4705.

Ford, P. J. (1982), Spin glasses, *Contemp. Phys.*, *23*, 141–168, doi:10.1080/00107518208237073.

Funabashi, K., and W. H. Hamill (1979), The role of electron-phonon interaction and non-Gaussian transport in spectral changes of trapped electrons in glasses, *Can. J. Chem.*, *57*, 197–206, doi:10.1139/v79-032.

Halgedahl, S. L. (1993), Experiments to investigate the origin of anomalously elevated unblocking temperatures, *J. Geophys. Res.*, *98*, 22,443–22,460, doi:10.1029/92JB02532.

Hamill, W. H., and K. Funabashi (1977), Kinetics of electron trapping reactions in amorphous solids; a non-Gaussian diffusion model, *Phys. Rev. B*, *16*, 5523, doi:10.1103/PhysRevB.16.5523.

Hara, H., and S. Okayama (1988), Fractal dimension and scaling behavior of cracks in a random medium: "Frequency-rank" distribution described by generalized random walks, *Phys. Rev. B*, *37*, 9504, doi:10.1103/PhysRevB.37.9504.

Hara, H., and Y. Tamura (2013), Dynamical process of complex systems and fractional differential equations, *Cent. Eur. J. Phys.*, *11*, 1238–1245, doi:10.2478/s11534-013-0224-2.

Kawada, Y., H. Nagahama, and H. Hara (2006), Irreversible thermodynamic and viscoelastic model for power-law relaxation and attenuation of rocks, *Tectonophysics*, *427*, 255–263, doi:10.1016/j.tecto.2006.03.049.

Kelso, P. R., and S. K. Banerjee (1994), Elevated temperature viscous remanent magnetization of natural and synthetic multidomain magnetite, *Earth Planet. Sci. Lett.*, *122*, 43–56, doi:10.1016/0012-821X(94)90050-7.

Kent, D. V. (1985), Thermoviscous remagnetization in some Appalachian limestones, *Geophys. Res. Lett.*, *12*, 805–808, doi:10.1029/GL012i012p00805.

Kohlrausch, R. (1854), Theorie des elektrischen ruckstandes in der leidner flasche, *Pogg. Ann. Phys.*, *91*, 179–214.

Kohlrausch, F. (1863), Ueber die elastische nachwirkung bei der torsion, *Pogg. Ann. Phys.*, *195*, 337–368, doi:10.1002/andp.18631950702.

Krause, S., G. Herzog, T. Stapelfeldt, L. Berbil-Bautista, M. Bode, E. Y. Vedmedenko, and R. Wiesendanger (2009), Magnetization reversal of nanoscale islands: How size and shape affect the Arrhenius prefactor, *Phys. Rev. Lett.*, *103*, 127202, doi:10.1103/PhysRevLett.103.127202.

Kubo, H., N. Nakamura, M. Kotsugi, T. Ohkochi, K. Terada, and K. Fukuda (2015), Striped domains of coarse-grained magnetite observed by X-ray photoemission electron microscopy as a source of the high remanence of granites in the Vredefort dome, *Front. Earth Sci.*, *3*, 31(1–8), doi:10.3389/feart.2015.00031.

Laherrère, J., and D. Sornette (1998), Stretched exponential distributions in nature and economy: "fat tails" with characteristic scales, *Eur. Phys. J. B*, *2*, 525–539, doi:10.1007/s100510050276.

Maxwell, J. C. (1867), On the dynamical theory of gases, *Phil. Trans. R. Soc. Lond.*, *157*, 49–88, doi:10.1098/rstl.1867.0004.
Middleton, M. F., and P. W. Schmidt (1982), Paleothermometry of the Sydney basin, *J. Geophys. Res.*, *87*, 5351–5359, doi:10.1029/JB087iB07p05351.
Moskowitz, B. M. (1985), Magnetic viscosity, diffusion after-effect, and disaccommodation in natural and synthetic samples, *Geophys. J. R. Astron. Soc.*, *82*, 143–161, doi:10.1111/j.1365-246X.1985.tb05133.x.
Muxworthy, A. R., J. Williams, and D. Heslop (2015), Testing the use of viscous remanent magnetisation to date flood events, *Front. Earth Sci.*, *3*, 1(1–9), doi:10.3389/feart.2015.00001.
Néel, L. (1949), Théorie du traînage magnétique des ferromagnétiques en grains fins avec applications aux terres cuites, *Ann. Géophys.*, *5*, 99–136.
Néel, L. (1955), Some theoretical aspects of rock-magnetism, *Adv. Phys.*, *4*, 191–243, doi:10.1080/00018735500101204.
Ngai, K. L. (1998), Relation between some secondary relaxations and the α relaxations in glass-forming materials according to the coupling model, *J. Chem. Phys.*, *109*, 6982–6994, doi:10.1063/1.477334.
Ngai, K. L. (2007), Why the glass transition problem remains unsolved? *J. Non-Cryst. Solids*, *353*, 709–718, doi:10.1016/j.jnoncrysol.2006.12.033.
Ngai, K. L., and U. Strom (1988), High-frequency dielectric loss of Na $\beta$-alumina: Evidence for relaxation crossover, *Phys. Rev. B*, *38*, 10,350, doi:10.1103/PhysRevB.38.10350.
Özdemir, Ö., and D. J. Dunlop (1999), Low temperature properties of a single crystal of magnetite oriented along principal magnetic axes, *Earth Planet. Sci. Lett.*, *165*, 229–239, doi:10.1016/S0012-821X(98)00269-6.
Ozima, M., M. Ozima, and T. Nagata (1964), Low-temperature treatment as an effective means of "magnetic cleaning" of natural remanent magnetization, *J. Geomagn. Geoelectr.*, *16*, 37–40, doi:10.5636/jgg.16.37.
Phillips, J. C. (1996), Stretched exponential relaxation in molecular and electronic glasses, *Rep. Prog. Phys.*, *59*, 1133–1207, doi:10.1088/0034-4885/59/9/003.
Plonka, A. (1983), Mechanism of hydrogen atom decay in γ-irradiated sulfuric acid glasses, *Radiat. Phys. Chem.*, *21*, 405–409, doi:10.1016/0146-5724(83)90019-5.
Plonka, A. (1986), *Time-Dependent Reactivity of Species in Condensed Media*, pp. 152, Springer, Berlin.
Porter, S. C., and T. Swanson (2008), $^{36}$Cl dating of the classic Pleistocene glacial record in the northeastern Cascade Range, Washington, *Am. J. Sci.*, *308*, 130–166, doi:10.2475/02.2008.02.
Porto, M. (2005), Ordered systems of ultrafine ferromagnetic particles, *Eur. Phys. J. B*, *45*, 369–375, doi:10.1140/epjb/e2005-00186-3.
Prester, M., I. Živković, D. Drobac, V. Šurija, D. Pajić, and H. Berger (2011), Slow magnetic dynamics and hysteresis loops of the bulk ferromagnet $Co_7(TeO_3)_4Br_6$, *Phys. Rev. B*, *84*, 064441, doi:10.1103/PhysRevB.84.064441.
Pullaiah, G., E. Irving, K. I. Buchan, and D. J. Dunlop (1975), Magnetization changes caused by burial and uplift, *Earth Planet. Sci. Lett.*, *28*, 133–143, doi:10.1016/0012-821X(75)90221-6.
Saito, R., and K. Murayama (1987), A universal distribution function of relaxation in amorphous materials, *Solid State Commun.*, *63*, 625–627, doi:10.1016/0038-1098(87)90867-2.
Sato, T., N. Nakamura, K. Goto, Y. Kumagai, H. Nagahama, and K. Minoura (2014), Paleomagnetism reveals the emplacement age of tsunamigenic coral boulders on Ishigaki Island, Japan, *Geology*, *42*, 603–606, doi:10.1130/G35366.1.
Scher, H., and E. W. Montroll (1975), Anomalous transit-time dispersion in amorphous solids, *Phys. Rev. B*, *12*, 2455, doi:10.1103/PhysRevB.12.2455.
Shlesinger, M. F., and B. D. Hughes (1981), Analogs of renormalization group transformations in random processes, *Phys. A: Stat. Mech. Appl.*, *109*, 597–608, doi:10.1016/0378-4371(81)90015-7.
Smith, B. M. (1984), Magnetic viscosity of some doleritic basalts in relation to the interpretation of the oceanic magnetic anomalies, *Geophys. Res. Lett.*, *11*, 213–216, doi:10.1029/GL011i003p00213.
Street, R., and J. C. Woolley (1949), A study of magnetic viscosity, *Proc. Phys. Soc. A*, *62*, 562–572, doi:10.1088/0370-1298/62/9/303.
Tivey, M., and H. P. Johnson (1984), The characterization of viscous remanent magnetization in large and small magnetite particles, *J. Geophys. Res.*, *89*, 543–552, doi:10.1029/JB089iB01p00543.
Trukhin, V. I., V. A. Zhilyaeva, A. I. Zhilyaeva, and I. A. Shubin (2003), Magnetic viscosity of basalts from fault zones of the Atlantic Ocean, *Izvestiya Phys. Solid Earth*, *39*, 666–675.
Tyson-Smith, R., and K. L. Verosub (1994), Thermoviscous remanent magnetism of Columbia River Basalt blocks in the Cascade landslide, *Geophys. Res. Lett.*, *21*, 2661–2664, doi:10.1029/94GL02669.
Ulrich, M., J. García-Otero, J. Rivas, and A. Bunde (2003), Slow relaxation in ferromagnetic nanoparticles: Indication of spin-glass behavior, *Phys. Rev. B*, *67*, 024416, doi:10.1103/PhysRevB.67.024416.
Walton, D. (1980), Time-temperature relations in the magnetization of assemblies of single domain grains, *Nature*, *286*, 245–247, doi:10.1038/286245a0.
Williams, G., and D. Watts (1970), Non-symmetrical dielectric relaxation behaviour arising from a simple empirical decay function, *Trans. Faraday Soc.*, *66*, 80–85, doi:10.1039/TF9706600080.
Williams, W., and A. R. Muxworthy (2006), Understanding viscous magnetization of multidomain magnetite, *J. Geophys. Res.*, *111*, B02102, doi:10.1029/2005JB003695.
Wohlfarth, E. P. (1977), Spin glasses exhibit rock magnetism, *Phys. B + C*, *86-88*, 852–853, doi:10.1016/0378-4363(77)90713-6.
Yu, Y., D. J. Dunlop, and Ö. Özdemir (2002), Partial anhysteretic remanent magnetization in magnetite 1. Additivity, *J. Geophys. Res.*, *107*(B10), 2244, doi:10.1029/2001JB001249.
Zapperi, S., P. Cizeau, G. Durin, and H. E. Stanley (1998), Dynamics of a ferromagnetic domain wall: Avalanches, depinning transition, and the Barkhausen effect, *Phys. Rev. B*, *58*, 6353, doi:10.1103/PhysRevB.58.6353.