

The effect of magnetic interactions on low temperature saturation remanence in fine magnetic particle systems

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In studies of fine magnetic particle systems, saturation remanence is often measured during warming from liquid helium temperature in order to determine the distribution of blocking temperatures. These data have usually been treated as if they are unaffected by magnetic interactions. However, this treatment is often inconsistent with the experimental data. Furthermore, the thermal decay of saturation remanence often gives values for the mean blocking temperature that are inconsistent with other measurements, such as low temperature ac susceptibility and zero-field-cooled magnetization curves. As an alternative interpretation of these remanence data, we suggest that interactions destabilize the saturation remanence state and accelerate its decay with increasing temperature. As a result, the blocking temperatures associated with the thermal decay of remanence are effectively reduced. We have modeled the effects of interactions on low temperature saturation remanence data using a simple mean interaction field model. This model produces remanence curves that have a steep slope at low temperatures, consistent with experimental curves frequently reported in the literature. © 2000 American Institute of Physics. [S0021-8979(00)05713-3]

I. INTRODUCTION

In most studies of low temperature saturation remanence in fine magnetic particle systems, these systems are treated as if interactions have no effect upon the saturation remanence (M_{rs}).¹⁻⁸ If these systems can be modeled as noninteracting single domain particles with a log-normal distribution of particle volumes, then M_{rs} , as a function of temperature T , should have zero slope at $T=0$; should decrease with increasing T ; and should have an inflection point where the rate of decrease reaches a maximum. However, experimental M_{rs} data (as a function of increasing T) frequently start with a rapid decrease at the lowest temperatures measured, and show no indication of an inflection point (see, for example, Fig. 1).¹⁻⁹ In Refs. 1, 2, and 3, theoretical fits have been made to M_{rs} vs T data, assuming noninteracting particles with a log-normal volume distribution, but these fits contain an inflection point that does not appear to be present in the data (see Fig. 1). Furthermore, in several of the same studies where a marked decrease in M_{rs} is observed at low T , M_{rs}/M_s has been extrapolated down to $T=0$ (where M_s is the saturation magnetization), and the result is less than the theoretical value expected for a system of noninteracting single domain particles.^{1,3,4,8,10}

Another problem with the conventional treatment of low temperature M_{rs} data is that, in several studies, mean blocking temperatures have been calculated from M_{rs} data which

are inconsistent with other measurements. In each case, mean blocking temperature obtained with M_{rs} data appears to be an underestimate. Five examples from the recent literature are given below.

(i) In studies of ferrofluids, El-Hilo *et al.*¹ obtained a mean blocking temperature of 32 K. According to their calculations, this implies that a peak should occur in the zero-field-cooled (ZFC) magnetization curve at 60 K. However, this predicted peak is 35 K lower than the actual measured peak for their most dilute sample (i.e., the sample with the least interactions).

(ii) In studies of barium ferrite particles, Batlle *et al.*⁴ obtained a mean blocking temperature of 81 K. According to their calculations, this gives a mean particle volume of $6 \times 10^4 \text{ \AA}^3$, which is less than the value obtained from transmission electron microscopy (TEM) observations ($1.1 \times 10^5 \text{ \AA}^3$).⁴

(iii) In studies of granular Ni-SiO₂ films, Xu *et al.*⁵ and Zhao *et al.*¹¹ used both remanence data and ac susceptibility data to measure mean blocking temperatures and to calculate mean particle volumes. However, the mean volume calculated from remanence data is less than the value calculated from the ac susceptibility data by a factor of roughly 5 (see footnote in Ref. 5).

(iv) In studies of nanosized maghemite particles, Morup *et al.*⁸ obtained a mean blocking temperature from M_{rs} data that is much lower than the same temperature obtained from Mössbauer spectroscopy and ZFC magnetization measurements.

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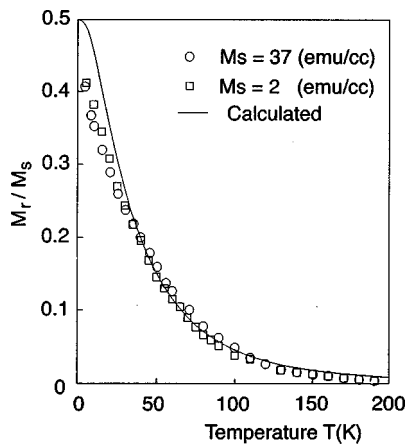


FIG. 1. The temperature variation of the reduced saturation remanence at different concentrations of fine Fe_2O_3 particles. The solid curve is the fit of a model assuming no interactions and a log-normal distribution of volumes [after El-Hilo *et al.* (Ref. 1)].

(v) In studies of iron oxyhydroxide particles, Dickson *et al.*⁷ obtained a mean blocking temperature of 9 K. From this they calculated the pre-exponential factor $f_0 \approx 10^{12}$ Hz. This is considerably larger than most experimental values (which would result from an underestimate of the mean blocking temperature).

In the above described examples, magnetic interactions are assumed to have no effect on the magnetic behavior at low temperatures. However, it has been suggested in the literature that magnetic interactions in fine magnetic particle systems will destabilize the saturation remanence state.^{8,10} In order to account for the above described data, we suggest the following hypothesis: at low T , these destabilizing interactions accelerate the decay of M_{rs} with increasing T ; this gives rise to an effective reduction in the blocking temperatures associated with saturation remanence. In this article, we discuss some possible objections to this hypothesis. We also model these destabilizing interactions with a simple mean field model. This model produces M_{rs} curves that have a steep slope at low T , consistent with the experimental curves frequently reported in the literature.

II. REDUCED BLOCKING TEMPERATURES

The effect of interactions in fine magnetic particle systems has been widely studied. It is widely thought that strong interactions at low temperatures will give rise to a spin-glass-like state.^{12–15} In low temperature ac susceptibility and ZFC magnetization measurements, an increase in particle concentration leads to increased blocking temperatures.^{13–15} At first glance, this result seems to contradict the above described hypothesis. However, no contradiction actually exists because the blocking temperatures obtained from M_{rs} data are not directly comparable to those obtained from ac susceptibility and ZFC magnetization measurements.

Both ac susceptibility and ZFC magnetization measurements are made with an applied field that is small enough that nonlinear effects can be ignored. In a ZFC magnetization measurement, the sample is cooled in zero field (starting from a high enough temperature that the sample is superpara-

magnetic) usually down to liquid helium temperature. A small applied field, usually not larger than 1 mT, is then applied and the magnetization is measured as a function of increasing temperature. In ac susceptibility measurements, a sample is also cooled in zero field, and an ac field usually not larger than 0.1 mT is applied. In these low field experiments, the low energy state of a fine magnetic particle system will consist of a spin-glass-like state with small net magnetization. Interactions act to stabilize this state, making it more difficult for an individual moment to switch direction; hence, interactions increase the blocking energies and temperatures. By contrast, in the low temperature saturation remanence state, the moments will be predominantly oriented towards the previously applied field. The same antiferromagnetic interactions which give rise to a spin-glass-like state can only push moments away from the direction of saturation remanence. Interactions will therefore destabilize the saturation remanence state, and decrease the blocking temperatures observed during thermal decay of saturation remanence. It is therefore possible that ac susceptibility and ZFC magnetization curves will yield different blocking temperatures than the thermal decay of saturation remanence.

It is more difficult to reconcile our hypothesis—that interactions reduce the blocking temperatures associated with the saturation remanence—with the results of El-Hilo *et al.*,¹ who report that M_{rs}/M_s is unaffected when the particle concentration is increased in a ferrofluid. An extrapolation of their M_{rs}/M_s data to $T=0$ actually appears to give a smaller remanence for the concentrated sample than for the dilute sample (Fig. 1). Nonetheless, the difference is small, and this has been cited as evidence that interactions do not affect M_{rs} .¹ This result, however, has another explanation: We suggest that in any ferrofluid, even a nominally dilute sample, there is inevitably a certain amount of particle agglomeration. In more concentrated samples, the distance between agglomerations decreases and their size increases, but small agglomerations are nonetheless present even in the most dilute samples. Djurberg *et al.*¹³ have found evidence for small particle agglomerations even in their most dilute ferrofluid, and even after the particles had been coated with a surfactant layer to prevent agglomeration. With regard to the data of El-Hilo *et al.*,¹ we suggest that most of the reduction in M_{rs} (due to interactions) takes place even in the dilute sample, due to small particle agglomerations.

In summary, our hypothesis—that interactions reduce the blocking temperatures associated with thermal decay of remanence—is consistent with experimental data. It is also consistent with a Monte Carlo simulation of an interacting granular magnetic solid at low temperatures.¹⁶

III. MEAN FIELD MODEL

We have modeled the effect of interactions on saturation remanence using a simple mean field treatment. A mean field treatment of interactions would not be applicable to a spin-glass-like state, where the moments order with no preferred direction. However, in a low temperature saturation remanence state, the moments will be predominantly oriented towards the previously applied field. Hence, the interaction field will also have a preferred direction. The fact that satu-

ration remanence is experimentally observed to be less than the noninteracting value indicates that the preferred direction of the interaction field is antiparallel to the magnetization. Our mean interaction field is intended to model this preferred direction. A mean field treatment of interactions in fine particle systems has previously proven useful in analyzing ferromagnetic resonance data¹⁷ and δM curves.¹⁸

Consider a collection of uniaxially anisotropic particles with identical volume V , anisotropy field H_k , and spontaneous magnetization I_s , where the particle easy axes are aligned with the z axis. Let the available orientation of each particle moment be restricted to the positive and negative z direction (up and down, respectively). Let $T_{\uparrow\downarrow}$ denote the probability per unit time that an up moment flips down; $T_{\downarrow\uparrow}$ is the opposite probability. Then

$$T_{\uparrow\downarrow} = f_a \exp(-EB_{\uparrow\downarrow}/kT), \tag{1}$$

where $EB_{\uparrow\downarrow}$ is the energy barrier that must be overcome for an up moment to switch down, k is Boltzmann's constant, and f_a is an "attempt" frequency, estimated at roughly 10^9 Hz. A similar expression applies to $T_{\downarrow\uparrow}$. The rate of change of the total normalized magnetic moment is

$$\dot{m} = -(T_{\uparrow\downarrow} - T_{\downarrow\uparrow}) - m(T_{\uparrow\downarrow} + T_{\downarrow\uparrow}). \tag{2}$$

In the presence of an applied field H_{app} , where $-H_k \leq H_{app} \leq H_k$, we have¹⁹

$$EB_{\uparrow\downarrow} = \frac{VI_s H_k}{2} (1 + H_{app}/H_k)^2 \tag{3}$$

and

$$EB_{\downarrow\uparrow} = EB_{\uparrow\downarrow} - 2VI_s H_{app}. \tag{4}$$

If $H_{app} < -H_k$ or $H_k < H_{app}$, then no energy barrier exists and all the moments align with the applied field. If we replace H_{app} with a mean interaction field $H_{mf} = -\alpha m H_k$, where α is dimensionless, then Eq. (2) becomes

$$\dot{m} = -f_a \exp\left[-\frac{VI_s H_k}{2kT} (1 + (\alpha m)^2)\right] \times \left(2 \sinh\left[\frac{VI_s H_k \alpha m}{kT}\right] + 2m \cosh\left[\frac{VI_s H_k \alpha m}{kT}\right]\right). \tag{5}$$

Consider next a collection of particles with a normalized distribution of volumes $F(V)$, where particles having volume between $V-dV$ and $V+dV$ make up a fraction $2dVF(V)$ of the total volume. Let us define a dimensionless volume $v \equiv V/\bar{V}$ and a dimensionless distribution $f(v) \equiv \bar{V}F(v\bar{V})$, where \bar{V} is the peak of $F(V)$. Let us write $M = \int dv f(v) m(v)$ and $H_{mf} = -\alpha M H_k$. Note that $f(v)$ will be peaked at $v = 1$. Let us also introduce a dimensionless temperature $\bar{T} \equiv kT/\bar{V}I_s H_k$. Then Eq. (5) becomes

$$\dot{m}(v, t) = -f_a \exp\left[-\frac{v}{2\bar{T}} (1 + \alpha^2 M(t)^2)\right] \times 2 \left(\sinh\left[\frac{v \alpha M(t)}{\bar{T}}\right] + m \cosh\left[\frac{v \alpha M(t)}{\bar{T}}\right]\right). \tag{6}$$

Equation (6) together with $M = \int dv f(v) m(v)$ comprises an integral differential equation for the time dependence of the magnetization.

In a measurement of M_{rs} , a saturating applied field is removed at $t=0$ and the magnetization is measured at a time t_m later. In a typical measurement, t_m is roughly 100 s. Henceforth in this article, we will fix t_m at 100 s, and M_{rs} will be treated as a function of \bar{T} .

To model the measurement of M_{rs} , for $\alpha < 1$, we have the initial condition $m(v, t=0) = M(t=0) = 1$. However, for $\alpha > 1$, if $M = 1$, this would make H_{mf} less than $-H_k$, which would imply that all the moments instantaneously switch to a negative orientation. Then, by the same reasoning, $M = -1$ and H_{mf} would be greater than H_k , which would imply that all the moments instantaneously switch to a positive orientation; this is a contradiction. The source of this problem is the assumption that the saturating field is removed instantaneously at $t=0$. In an actual measurement, H_{app} is ramped down from a large positive value to zero over some finite time interval (of length Δt) ending at $t=0$. As H_{app} is ramped down, M must decrease enough that the above described contradiction is avoided, i.e., $H_{app} + H_{mf} \geq -H_k$. Inserting, $H_{mf} = -\alpha M H_k$, this becomes

$$M \leq (H_k + H_{app})/(H_k \alpha). \tag{7}$$

While H_{app} is ramping down, $M(t)$ will be governed by Eq. (6) but is also constrained by the upper bound in Eq. (7) and by $M \leq 1$. In the limit where Δt is infinitesimally small, Eq. (6) can lead to only an infinitesimal decrease in $M(t)$ as the field is ramped down. Therefore, in this limit, M approaches the lesser of the two upper bounds. When $t=0$, we have $H_{app}=0$, and Eq. (A27) becomes $M \leq 1/\alpha$. For $\alpha > 1$, this becomes the dominant upper bound. Therefore, in the limit of small Δt , for $\alpha > 1$, the initial condition becomes $M(t=0) = 1/\alpha$.

IV. MODEL RESULTS

Analytical solutions to the mean field model can be obtained in certain limiting cases, as described in the appendices. In the limit of small α (Appendix A), the mean interaction field shifts the peak in the distribution of blocking temperatures, $\rho(\bar{T}) = -dM_{rs}/d(\bar{T})$, to lower \bar{T} (Fig. 2). (As shown in Appendix A, the unblocking temperature of a non-interacting particle with $v=1$ is $\bar{T}=1/C$, where $C \equiv 2 \ln[2t_m f_a / \ln(2)]$). Therefore, it is natural to plot our results as a function of $C\bar{T}$. For $\alpha \geq 0$, in the limit as \bar{T} goes to zero (Appendix B), $M(\bar{T})$ approaches 1, and $\rho(\bar{T})$ approaches zero; this implies that a peak is still present in the distribution of blocking temperatures at some $T > 0$. For $\alpha \geq 1$, in the limit as \bar{T} and Δt go to zero (Appendix C), we have simpli-

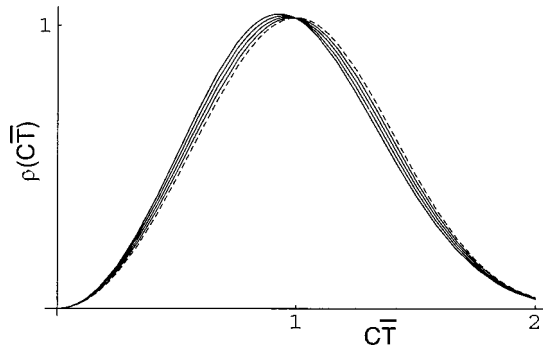


FIG. 2. The distribution of blocking temperatures $\rho(C\bar{T})$ as a function of $C\bar{T}$, calculated using $f(v)=2v^2\exp(-0.666669v^3)$ and Eqs. (A14) and (A15), for $\alpha C=[0,0.07,0.14,0.21,0.28]$. The dotted line is the noninteracting case, i.e., $\alpha C=0$. Curves for increasing values of αC have peaks which progress to lower $C\bar{T}$. We defined \bar{T} so that a particle at the peak volume, \bar{V} , has an unblocking temperature $\bar{T}=1/C$, i.e., $C\bar{T}=1$.

fied the model by letting all the particles have identical volume, and we have shown that M_{rs} has the functional form $(1/\alpha)(1-a\sqrt{\bar{T}})$, where a is a fitting parameter. Hence, the peak in the distribution of blocking temperatures has vanished. Zhao *et al.* found that their low temperature M_{rs} data fit this functional form.¹¹

In our numerical calculations, the continuous volume distribution was approximated by delta functions at ten equally spaced volumes v_i . The model then becomes a system of ten coupled differential equations that can be solved by the Runge-Kutta method. These delta functions were each weighted by $f(v_i)$, where $f(v)$ is a log-normal distribution with a peak at 1 and logarithmic standard deviation of 0.26. To obtain M_{rs} for $\alpha \leq 1$, Eq. (6) was evaluated at $t_m = 100$ s, with initial condition $m(t=0, v_i) = 0$. For $\alpha > 1$, we

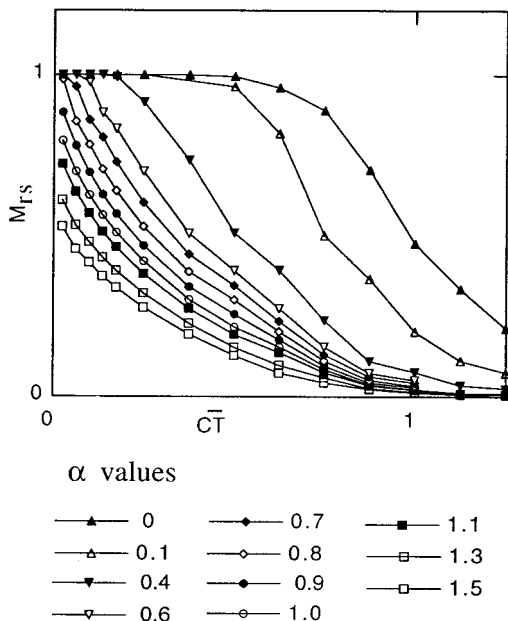


FIG. 3. Numerical calculation of saturation remanence as a function of $C\bar{T}$ for $0 \leq \alpha \leq 1.5$. For $f(v)$, a log-normal distribution was used with a peak at $v=1$ and logarithmic standard deviation of 0.26. With increased interactions, the effective blocking temperatures progressively decrease.

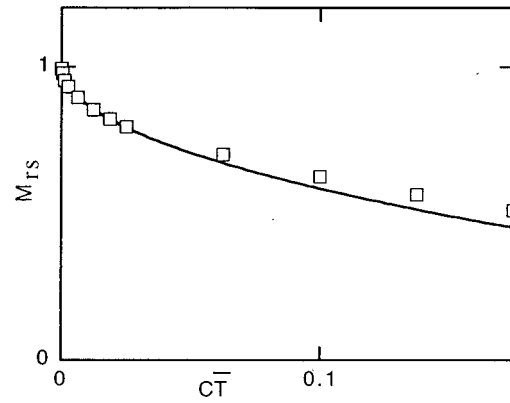


FIG. 4. Additional calculations of saturation remanence at lower \bar{T} for $\alpha = 1$. The same function was used as for $f(v)$ as in Fig. 3. The lowest 8 points in \bar{T} were fitted to $(1-a\sqrt{\bar{T}})$, where $a=1.3$. These data illustrate the marked thermal decay of M_{rs} that is often observed in experimental results.

incorporated an applied field that starts at a large positive value (just prior to $t=0$) and is quickly ramped down to zero (at $t=0$).

Calculations of M_{rs} are shown as a function of $C\bar{T}$ for a range of α values in Fig. 3. Additional low temperature points are shown for $\alpha=1$ in Fig. 4; note that M_{rs} appears to approach 1 as \bar{T} goes to zero for $\alpha=1$. In Fig. 3, for $\alpha > 1$, M_{rs} appears to approach a value less than 1 as \bar{T} goes to zero. These numerical results also indicate that when interactions are strong (i.e., $\alpha \geq 1$), M_{rs} has a steep slope at low \bar{T} . This is consistent with the low T remanence curves frequently found in the literature¹⁻⁹ and suggests that interactions should be considered as an explanation for curves with this behavior. The eight points with lowest \bar{T} in Fig. 4 closely fit the function $(1-1.3\sqrt{\bar{T}})$. This functional form is consistent with our analytical results and also with the experimental data of Zhao *et al.*¹¹

V. CONCLUSIONS

In the presence of strong magnetic interactions, the distribution of blocking temperatures associated with the thermal decay of saturation remanence is not directly comparable to the distribution of blocking temperatures obtained from ac susceptibility or ZFC magnetization curves. This is because the later are measured in low fields, in which these systems order into spin-glass-like states that are stabilized by interactions, leading to increased blocking temperatures. By contrast, interactions destabilize the saturation remanence state, drive it to a lower magnetization, and lead to an apparent reduction in the blocking temperatures. A mean interaction field model predicts that when interactions are strong, there will be a reduction in M_{rs} at $T=0$, and that M_{rs} will have a $(1-a\sqrt{\bar{T}})$ functional dependence at low T , where a is a fitting parameter. This is consistent with the marked decrease in M_{rs} that is observed with increasing T in many fine magnetic particle systems. It also suggests that the effects of magnetic interactions have been inappropriately ignored in many studies of low temperature saturation rema-

nence. Pike *et al.*²⁰ recently described a sensitive technique for estimating the effects of magnetic interactions in fine particle systems. Use of this technique at low T might help to resolve some of the anomalies that are frequently seen in analyses of low temperature M_{rs} data.

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APPENDIX A: ANALYTIC SOLUTION FOR SMALL α

We are interested in the effect of weak interactions on the peak of the distribution of blocking temperatures, $\rho(\bar{T})$. Let us treat this as a perturbation of the noninteracting case, i.e., $\alpha=0$. We will let $m_o(v,t)$ denote the solution of Eq. (6) for $\alpha=0$, with initial condition $m_o(v,t=0)=1$. It can be shown that:

$$m_o(v,t) = \exp[-2tf_a\Gamma(v,\bar{T})], \tag{A1}$$

where we use the notation $\Gamma(v,\bar{T}) \equiv \exp[-v/2\bar{T}]$. Similarly, the total magnetization in the noninteracting case will be denoted by $M_o(t)$, and this equals

$$M_o(t) = \int dvf(v)\exp[-tf_a2\Gamma(v,\bar{T})]. \tag{A2}$$

It is well known that, due to the large magnitude of $(t_m f_a)$, $\exp[-t_m f_a 2\Gamma(v,\bar{T})]$ can be approximated by the step function $\Theta(v-v_o^{ct})$, where $\Theta(x) = \{0 \text{ for } x < 0; 1 \text{ for } x > 0\}$, where v_o^{ct} is a cutoff volume given by

$$v_o^{ct} = C\bar{T}, \tag{A3}$$

where $C \equiv 2 \ln[2t_m f_a / \ln(2)]$; the subscript ‘‘o’’ again indicates this is the noninteracting system. Thus, in the noninteracting case

$$M_{rs}(\bar{T}) \equiv M(t_m) = 1 - \int_0^{v_o^{ct}} dvf(v). \tag{A4}$$

Equation (A4) implies that the peak of $\rho(\bar{T})$ occurs when v_o^{ct} is at the peak of $f(v)$, i.e., when $v_o^{ct} = 1$. Hence, the peak of $\rho(\bar{T})$ is at $\bar{T} = 1/C$. The following perturbation analysis will be carried out for v near 1 and \bar{T} near $1/C$.

Let us expand Eq. (6) to first order in α :

$$\dot{m}(v,t) = -2f_a\Gamma(v,\bar{T})(m(v,t) + \alpha v M(t)/\bar{T}). \tag{A5}$$

We can write $m(v,t) = m_o(v,t) + \alpha m_1(v,t)$, and where $m_1(v,t=0) = 0$. Similarly, we write $M(t) = M_o(t) + \alpha M_1(t)$. Collecting terms to first order in α , Eq. (A5) becomes

$$\begin{aligned} \dot{m}_o(v,t) + \alpha \dot{m}_1(v,t) = & -2f_a\Gamma(v,\bar{T}) \left(\dot{m}_o(v,t) \right. \\ & \left. + \alpha \dot{m}_1(v,t) + \alpha M_o(t) \frac{v}{\bar{T}} \right). \end{aligned} \tag{A6}$$

Because $m_o(v,t)$ solves Eq. (6) with $\alpha=0$, Eq. (A6) becomes

$$\dot{m}_1(v,t) = -2f_a\Gamma(v,\bar{T}) \left(m_1(v,t) + M_o(t) \frac{v}{\bar{T}} \right). \tag{A7}$$

Equation (A7) with initial condition $m_1(v,t=0) = 0$ has the solution

$$\begin{aligned} m_1(v,t) = & -\frac{2vf_a}{\bar{T}}\Gamma(v,\bar{T}) \int_0^t dt' M_o(t') \\ & \times \exp[-2(t-t')f_a\Gamma(v,\bar{T})]. \end{aligned} \tag{A8}$$

Inserting Eq. (A2) for $M(t')$, with ν for the dummy variable in the integral, Eq. (A8) becomes

$$\begin{aligned} m_1(v,t) = & \exp[-2tf_a\Gamma(v,\bar{T})] \\ & \times \left(-\frac{2vf_a}{\bar{T}} \right) \Gamma(v,\bar{T}) \int dvf(\nu) \int_0^t dt' \\ & \times \exp[2t'f_a\Gamma(v,\bar{T})] \exp[-2t'f_a\Gamma(\nu,\bar{T})]. \end{aligned} \tag{A9}$$

Evaluating Eq. (A9) at $t=t_m$ gives

$$\begin{aligned} m_1(v,t_m) & = \exp[-2t_m f_a \Gamma(v,\bar{T})] \left(-\frac{2vf_a}{\bar{T}} \right) \Gamma(v,\bar{T}) \int dvf(\nu) \\ & \times \left[\frac{\exp[2t_m f_a (\Gamma(v,\bar{T}) - \Gamma(\nu,\bar{T}))] - 1}{2f_a (\Gamma(v,\bar{T}) - \Gamma(\nu,\bar{T}))} \right] \\ & = -\frac{v}{\bar{T}} \int dvf(\nu) \\ & \times \left\{ \frac{\exp[-2t_m f_a \Gamma(v,\bar{T})] - \exp[-2t_m f_a \Gamma(\nu,\bar{T})]}{\Gamma(\nu - v, \bar{T}) - 1} \right\}. \end{aligned} \tag{A10}$$

It can be shown numerically that, for \bar{T} near $1/C$ and v near 1, the expression in the curly brackets of Eq. (A10), as a function of ν , can be approximated by this step function:

$$\begin{aligned} \{ \dots \text{curly brackets in Eq. (A10)} \dots \} \\ = (1 - \exp[-2t_m f_a \Gamma(v,T)]) \theta(\nu - v_o^{ct}). \end{aligned} \tag{A11}$$

The coefficient in front of $\theta(\nu - v_o^{ct})$ in Eq. (A11) can be obtained by taking the limit of the expression in the curly brackets as ν goes to infinity. But $\exp[-2t_m f_a \Gamma(v,T)]$ is just $m_o(v,t_m)$ and can be approximated by the step function $\theta(v - v_o^{ct})$. So,

$$\begin{aligned} \{ \dots \text{curly brackets in Eq. (A10)} \dots \} \\ = (1 - \theta(v - v_o^{ct})) \theta(\nu - v_o^{ct}). \end{aligned} \tag{A12}$$

Integrating over ν in Eq. (A10) gives

$$\begin{aligned}
 m_1(v, t_m) &= -\frac{v}{\bar{T}}(1 - \theta(v - v_o^{ct})) \int_0^\infty dv f(v) \theta(v - v_o^{ct}) \\
 &= -\frac{v}{\bar{T}}(1 - \theta(v - v_o^{ct})) \int_{v_o^{ct}}^\infty dv f(v). \tag{A13}
 \end{aligned}$$

We next insert Eq. (A13) into $m(v, t_m) = m_o(v, t_m) + \alpha m_1(v, t_m)$, and insert this into $M = \int dv f(v) m(v)$. We replace $m_o(v, t_m)$ with $\theta(v - v_o^{ct})$. We get

$$M_{rs} = M(t_m) = \left(\int_{C\bar{T}}^\infty dv f(v) \right) \left(1 - \frac{(\alpha C)}{C\bar{T}} \int_0^{C\bar{T}} dv \times v f(v) \right). \tag{A14}$$

Equation (A14) reduces to Eq. (A4) for the noninteracting case. The distribution of blocking temperatures as a function of CT becomes:

$$\rho(C\bar{T}) = -\frac{dM_{rs}}{d(C\bar{T})}. \tag{A15}$$

To proceed further, we insert a specific function for $f(v)$ into Eq. (A14). If a log-normal function is used for $f(v)$, then the indefinite integral of $vf(v)$ does not have a closed form. We have used the distribution $f(v) = 2v^2 \exp \times (-0.66667v^3)$, which is peaked at $v = 1$, in order to obtain a closed form solution. Kaysser *et al.* derived this size distribution from a model of particle growth by coalescence during liquid phase sintering.²¹ We have evaluated $\rho(C\bar{T})$ for several values of $(C\alpha)$ using Eqs. (A14) and (A15). The results are shown in Fig. 2.

APPENDIX B: ANALYTIC SOLUTION FOR SMALL \bar{T} ; $\alpha < 1$

In the limit as \bar{T} goes to zero, Eq. (6) becomes

$$\begin{aligned}
 \dot{m}(v, t) &= -f_a \exp \left[-\frac{v}{2\bar{T}}(1 + \alpha^2 M(t)^2) \right] \\
 &\times (1 + m(v, t)) \exp \left[\frac{v \alpha M(t)}{\bar{T}} \right] \\
 &= -f_a \exp \left[-\frac{v}{2\bar{T}}(1 - \alpha M(t))^2 \right] (1 + m(v, t)). \tag{A16}
 \end{aligned}$$

The solution of Eq. (A16) with initial condition $m(v, t=0) = 1$ is

$$\begin{aligned}
 m(v, t) &= 2 \exp \left[-f_a \int_0^t dt' \right. \\
 &\left. \times \exp \left[-\frac{v}{2\bar{T}}(1 - \alpha M(t'))^2 \right] \right] - 1. \tag{A17}
 \end{aligned}$$

Because $\alpha < 1$ and $M \leq 1$, $m(v, t)$ given by Eq. (A16) will go to 1 in the limit as \bar{T} goes to zero. Hence, $M(t)$ and $M_{rs}(\bar{T})$ also go to 1. Therefore, in the limit as \bar{T} goes to zero, $[1 - M(t)]$ goes to zero, and we can write

$$\begin{aligned}
 \text{Limit}_{\bar{T} \rightarrow 0} (1 - \alpha M(t))^2 &= (1 - \alpha)^2 \\
 &+ \text{Limit}_{\bar{T} \rightarrow 0} \{ -2\alpha(1 - \alpha)[M(t) - 1] + \alpha^2[M(t) - 1]^2 \} \\
 &= (1 - \alpha)^2 + 2\alpha(1 - \alpha)(1 - M(t)). \tag{A18}
 \end{aligned}$$

Thus, in the limit as \bar{T} goes to zero, we can write Eq. (A17) as

$$\begin{aligned}
 m(v, t) &= 2 \exp \left[-f_a \exp \left[-\frac{v}{2\bar{T}}(1 - \alpha)^2 \right] \int_0^t dt' \right. \\
 &\left. \times \exp \left[-\frac{v}{2\bar{T}} 2\alpha(1 - \alpha)(1 - M(t')) \right] \right] - 1. \tag{A19}
 \end{aligned}$$

Since $[1 - M(t')]$, $(1 - \alpha)$, and α are greater than zero, then $t > \int_0^t dt' \exp[-v/2\bar{T} 2\alpha(1 - \alpha)(1 - M(t'))]$.

We can insert this inequality into Eq. (A19) to get

$$m(v, t) > 2 \exp \left[-f_a t \exp \left[-\frac{v}{2\bar{T}}(1 - \alpha)^2 \right] \right] - 1. \tag{A20}$$

If we insert $m(v, t)$ into $M = \int dv f(v) m(v)$ and evaluate M at $t = t_m$, this gives us $M_{rs}(\bar{T})$. Using the inequality of Eq. (A20) gives

$$\begin{aligned}
 1 - M_{rs}(\bar{T}) &< 2 - \int dv f(v) 2 \exp \left[-f_a t_m \right. \\
 &\left. \times \exp \left[-\frac{v}{2\bar{T}}(1 - \alpha)^2 \right] \right]. \tag{A21}
 \end{aligned}$$

The exponent in Eq. (A21) [compare with Eq. (A1)] can be approximated by the step function $\theta(v - v_o^{ct}/(1 - \alpha)^2)$. Assuming $f(v)$ is a slowly varying function of v and inserting $v_o^{ct} = C\bar{T}$, Eq. (A21) becomes

$$1 - M_{rs}(\bar{T}) < 2 \int_0^{C\bar{T}/(1 - \alpha)^2} dv f(v). \tag{A22}$$

Using Eq. (A22), the distribution of blocking temperatures at $\bar{T} = 0$ can be written

$$\begin{aligned}
 \rho(0) &\equiv \text{Limit}_{\bar{T} \rightarrow 0} \frac{-(M_{rs}(\bar{T}) - M_{rs}(\bar{T}=0))}{\bar{T}} \\
 &< \text{Limit}_{\bar{T} \rightarrow 0} \frac{2}{\bar{T}} \int_0^{C\bar{T}/(1 - \alpha)^2} dv f(v), \tag{A23}
 \end{aligned}$$

where $M_{rs}(\bar{T}=0) = 1$ for $\alpha < 1$. We know that $f(v)$ vanishes at $v = 0$, and we can assume that it is positive and well-behaved for $v > 0$, so we can write

$$\text{Limit}_{x \rightarrow 0} \frac{1}{x} \int_0^x dv f(v) < f(x), \tag{A24}$$

and Eq. (A23) becomes

$$\rho(\bar{T}=0) < \frac{2C}{(1-\alpha)^2} f\left(\frac{C\bar{T}}{(1-\alpha)^2}\right). \tag{A25}$$

Hence, $\rho(\bar{T})$ goes to zero in the limit as \bar{T} goes to zero.

APPENDIX C: ANALYTICAL SOLUTION $\alpha \geq 1$ AND SMALL \bar{T}

Let us simplify the model by letting all particles have identical volume, so $v = 1$, and $M = m(v = 1)$. In the limit of small \bar{T} , Eq. (6) becomes Eq. (A16). With initial condition $M(t=0) = 1/\alpha$ this has solution

$$M(t, \bar{T}) = \left(1 + \frac{1}{\alpha}\right) \exp\left[-f_a \int_0^t dt'\right] \times \exp\left[-\frac{1}{2\bar{T}}(1 - \alpha M(t', \bar{T}))^2\right] - 1. \tag{A26}$$

Let us define $\phi(t, \bar{T})$ such that

$$M(t, \bar{T}) = (1/\alpha)[1 - \sqrt{\bar{T}}\phi(t, \bar{T})]. \tag{A27}$$

By inspection of Eq. (A26) we know that $M(t, \bar{T})$ is less than $1/\alpha$ and that, at fixed t , $M(t, \bar{T})$ goes to $1/\alpha$ as \bar{T} goes to 0. Therefore, $\phi(t, \bar{T})$ is a positive function and, at a fixed t , $\sqrt{\bar{T}}\phi(t, \bar{T})$ goes to 0 as \bar{T} goes to 0. We will next prove that, keeping t fixed, as \bar{T} goes to 0, $\phi(t, \bar{T})$ goes to infinity, but it does so slower than $(1/\bar{T})^\gamma$ for any positive number γ . In other words, as \bar{T} goes to 0, the order of $\phi(t, \bar{T})$ in $(1/\bar{T})$ approaches zero. This implies that $M(t, \bar{T})$ approaches $(1/\alpha)[1 - a\sqrt{\bar{T}}]$, where a is a fitting parameter.

Proof:

Inserting Eq. (A27) into Eq. (A26) gives

$$\frac{1 - \sqrt{\bar{T}}\phi(t, \bar{T}) + \alpha}{1 + \alpha} = \exp\left[-f_a \int_0^t dt' \exp\left[-\frac{\phi(t', \bar{T})^2}{2}\right]\right]. \tag{A28}$$

Let t^* be a fixed time, and let t_1 and t_2 be two times such that $t_1 \leq t^* \leq t_2$, where $(t_2 - t_1)$ is small. Let us evaluate Eq. (A28) at t_1 and t_2 , and divide the later result by the former. We get

$$\frac{1 - \sqrt{\bar{T}}\phi(t_2, \bar{T}) + \alpha}{1 - \sqrt{\bar{T}}\phi(t_1, \bar{T}) + \alpha} = \exp\left\{-f_a \int_{t_1}^{t_2} dt' \exp\left[-\frac{\phi(t^* \bar{T})^2}{2}\right]\right\}. \tag{A29}$$

Since $(t_2 - t_1)$ is small, we can write, Eq. (A29) as

$$1 - \frac{\sqrt{\bar{T}}(t_2 - t_1) \frac{\partial}{\partial t} \phi(t, \bar{T}) \Big|_{t^*}}{1 - \sqrt{\bar{T}}\phi(t_1, \bar{T}) + \alpha} = \exp\left\{-f_a(t_2 - t_1) \exp\left[-\frac{\phi(t^*, \bar{T})^2}{2}\right]\right\}. \tag{A30}$$

Since $\sqrt{\bar{T}}\phi(t^*, \bar{T})$ goes to 0 as \bar{T} goes to 0, and since $\sqrt{\bar{T}}\phi(t, \bar{T})$ is positive, we know that $\sqrt{\bar{T}}\partial/\partial t[\phi(t, \bar{T})]_{t^*}$ must also go to 0. Therefore, as \bar{T} goes to 0, the left hand side of Eq. (A30) goes to 1. In order that the right hand side also goes to 1, we know that $\phi(t^*, \bar{T})$ must go to infinity.

Since both sides of Eq. (A30) go to 1 as \bar{T} goes to 0, we can expand the exponent on the right hand side to first order of its argument. Let us also subtract one from each side of Eq. (A30), and multiply by sides by a negative sign, and omit $\sqrt{\bar{T}}\phi(t_1, \bar{T})$ in the denominator of the left hand side, since this goes to 0 as \bar{T} goes to 0. Then, in the limit as \bar{T} goes to 0, Eq. (A30) becomes

$$\frac{\sqrt{\bar{T}}(t_2 - t_1) \frac{\partial}{\partial t} \phi(t, \bar{T}) \Big|_{t^*}}{1 + \alpha} = f_a(t_2 - t_1) \exp\left[-\frac{\phi(t^*, \bar{T})^2}{2}\right]. \tag{A31}$$

Since $\phi(t, \bar{T})$ is a positive function, then in the limit as \bar{T} goes to 0, we know that $\partial\phi(t, \bar{T})/\partial t|_{t^*}$ will have the same order in $(1/\bar{T})$ as does $\phi(t^*, \bar{T})$. Therefore, Eq. (A31) implies

$$O_{(1/\bar{T})}(\sqrt{\bar{T}}\phi(t^*, \bar{T})) = O_{(1/\bar{T})}\left(\exp\left[-\frac{1}{2}\phi(t^*, \bar{T})^2\right]\right), \tag{A32}$$

where $O_{(1/\bar{T})}$ gives the order in $(1/\bar{T})$ of the indicated expression. Let us next suppose that in the limit as \bar{T} goes to 0, $\phi(t^*, \bar{T}) > (1/\bar{T})^\gamma$ for some positive number γ . Then

$$O_{(1/\bar{T})}(\sqrt{\bar{T}}\phi(t^*, \bar{T})) > \gamma - 1/2, \tag{A33}$$

and also

$$O_{(1/\bar{T})}\left(\exp\left[\frac{1}{2}(1/\bar{T})^{2\gamma}\right]\right) < O_{(1/\bar{T})}\left(\exp\left[\frac{1}{2}\phi(t^*, \bar{T})^2\right]\right) = -O_{(1/\bar{T})}\left(\exp\left[-\frac{1}{2}\phi(t^*, \bar{T})^2\right]\right). \tag{A34}$$

Combining Eqs. (A32)–(A34), we get

$$O_{(1/\bar{T})}\left(\exp\left[\frac{1}{2}(1/\bar{T})^{2\gamma}\right]\right) < 1/2 - \gamma. \tag{A35}$$

But, in the limit as \bar{T} goes to 0, the exponent in Eq. (A34) will have a larger order in $(1/\bar{T})$ than any polynomial. Therefore, a contradiction has occurred. And therefore we know that in the limit as \bar{T} goes to 0, $\phi(t^*, \bar{T})$ will be smaller than $(1/\bar{T})^\gamma$ for any positive value of γ .

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