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WORK PROBABILITY DISTRIBUTION FOR A FERROMAGNET WITH LONG-RANGED AND SHORT- RANGED CORRELATIONS

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ABSTRACT

Work fluctuations and work probability distributions are fundamentally different in systems with short- ranged versus long-ranged correlations. Specifically, in systems with long-ranged correlations the work distribution is extraordinarily broad compared to systems with short-ranged correlations. This difference profoundly affects the possible applicability of fluctuation theorems like the Jarzynski fluctuation theorem. The Heisenberg ferromagnet , well below its Curie temperature, is a system with long-ranged correlations in very low magnetic fields due to the presence of Goldstone modes. As the magnetic field is increased the correlations gradually become short-ranged. Hence, such a ferromagnet is an ideal system for elucidating the changes of the work probability distribution as one goes from a domain with long-ranged correlations to a domain with short-ranged correlations by tuning the magnetic field. A quantitative analysis of this crossover behaviour of the work probability distribution and the associated fluctuations is presented.

I **INTRODUCTION**

One of the most significant developments in non-equilibrium statistical mechanics in the last few decades has been the emergence of a set of fluctuation theorems [1-6], some of which [4-6] concern the fluctuations in the work done as a system moves from an initial thermodynamic state at temperature T to a final thermodynamic state at the same temperature. The passage from the initial to the final state can occur along any one of the infinitely available paths. Hence one needs to average over an ensemble of paths, when one considers any function of the work (W) done in the transition from the initial to the final state. A primary example of such a fluctuation- dissipation theorem is Jarzynski's equality [4] which yields

$$< e^{-\frac{W}{k_B T}} >= e^{-\frac{\Delta F}{k_B T}} \qquad (1.1)$$

1

where W is the work done in going from state 1 (initial) to state 2 (final), k_B is Boltzmann's constant and where $\Delta F = F_2 - F_1$, with F_1 and F_2 being the thermodynamic free energies (Helmholtz energies) associated with states 1 and 2, respectively.

The appearance of an expectation value on the left- hand side of Eq. (1.1) raises questions about the probability distribution of the work fluctuations. The fact that extreme events can play a role in determining the possible non-Gaussian tail of the distribution has motivated researchers to find the distribution both theoretically and experimentally [7-11]. Crooks and Jarzynski [12] have considered a particularly illuminating example involving the adiabatic compression of an ideal monatomic gas from an initial volume V_0 to a final volume V_1. In terms of a parameter α defined by the relation $1 + \alpha = \left(V_0 / V_1 \right)^{2/3}$; they found for the work probability distribution $\rho(W)$ in this case (note that $W > 0$ in this case)

$$\rho(W) = \frac{1}{\alpha k_B T \Gamma(3N/2)} \left(\frac{W}{\alpha k_B T} \right)^{\frac{3N}{2} - 1} e^{-\frac{W}{\alpha k_B T}} \qquad (1.2)$$

where N is the number of molecules of the gas.

As noted recently by Kirkpatrick $et\ al.$ [13], this distribution is centered at $\langle W \rangle = 3N\alpha / 2$ and around the centre can be approximated by the Gaussian form

$$\rho_{\text{approx}}(W) \propto e^{-\frac{3N}{4} \left(\frac{W}{\langle W \rangle} - 1 \right)^2} \qquad (1.3)$$

With the distribution peaked at $\langle W \rangle$ which is $O(N)$ and with a width which is $O(\sqrt{N})$, $\rho(W)$ is sharply peaked at its maximum for $N \gg 1$. Around the maximum of the distribution, the function $\Omega = \exp(-W / k_B T)$ is already very small and hence fluctuations in it can be significant. The root- mean- square fluctuation ε_Ω of Ω is defined as

$$\varepsilon_\Omega = \frac{\langle \Omega^2 \rangle}{\langle \Omega \rangle^2} - 1 = \frac{\langle \exp(-2W / k_B T) \rangle}{\langle \exp - W / k_B T \rangle^2} - 1 \qquad (1.4)$$

Using the distribution, given by Eq. (1.2), we get ,

$$\varepsilon_\Omega = \exp[\frac{3N}{2} \ln(1 + \frac{\alpha^2}{1 + 2\alpha})] \qquad (1.5)$$

What is striking about the above equation is that the fluctuation is exponentially large in the system size N and thus makes the usefulness of the equality shown in Eq. (1.1) , which is formally correct, questionable. This will become a problem for large systems where $N \gg 1$

2

and ε_Ω needs to be vanishingly small if the system is supposed to be well -behaved. Instead we have in this case exponentially large (in the system size) fluctuations around the mean. Even for systems where N is of $O(1)$, the fluctuations are of the same order as the mean, which makes the approach to the mean value difficult. The Gaussian approximation of Eq. (1.4) yields $\varepsilon_\Omega = \exp[3N\alpha^2/2]$, which agrees with the result obtained from the exact distribution for $\alpha << 1$ as expected. The difficulty with an experimental implementation of the Jarzynski equality has been carefully examined by Suarez *et al.* [14]. These authors have found the lower bound M_c on the number of times an experiment must be repeated to faithfully construct the average value shown in Eq. (1.1). For N particles, the number M_c was found to be of $O(e^N)$, consistent with our measure of fluctuation around the mean. The difficulty in approaching the mean in numerical works have been pointed out in references [15-18]. These very large fluctuations result from the short- range nature of the correlations in the systems considered. This feature can also be seen in a situation with a set of independent oscillators envisaged by Hijar and Ortiz de Zárate[19].

We now discuss what happens if the correlations are of a long- ranged nature. As shown in a series of papers in the case of a fluid under a fixed temperature gradient [20-23], one has a non-equilibrium steady state (NESS) with generic long- range correlations. These long-ranged correlations are capable of producing large Casimir type forces in confined fluid layers [24-27]. Recently it was found that the work probability distribution in such cases can be significantly different [13]. This prompted us to look at systems with long-ranged correlations in both equilibrium and non-equilibrium situations [28]. A natural candidate for an equilibrium system was a Heisenberg ferromagnet, well below the Curie temperature, in an infinitesimal magnetic field, where the transverse magnetization fluctuations are the Goldstone modes of the system. These modes , being massless , lead to the fluctuations having long- ranged correlations. As anticipated, the long- ranged correlation between the fluctuations lead to a work probability distribution similar to the case of NESS with generic long-ranged fluctuation correlations. The important point about the probability distributions in the case of systems with these long-ranged correlations is that the distribution is wider by orders of magnitude and this greatly reduces the fluctuations ε_Ω around $\left\langle \exp - (W/k_BT) \right\rangle$.

The Heisenberg ferromagnet at temperatures well below the Curie temperature actually provides a single system where both short- ranged and long- ranged correlations can be probed. Specifically, one can use the external magnetic field as a tuning parameter to go from long-ranged to short-ranged fluctuations. Consequently, this is an ideal testing ground for studying the crossover in the probability distribution of the work and in the fluctuation ε_Ω , as one goes from long- ranged correlations to short- ranged ones.

3

The factors of *N* in Eqs. (1.2), (1.3) and (1.5) are directly related to the fact that the root-mean-squared fluctuations in a system with short- ranged correlations scale as $1/\sqrt{N}$. The work distribution function and ε_Ω are fundamentally different when there are long- ranged correlations. For example in the case considered here the local work distribution function in three dimensions has correlations that decay as the square of the distance for small magnetic fields. This in turn determines that the total work fluctuations scale as

$$\frac{\left\langle (\delta W)^2 \right\rangle}{\left\langle W \right\rangle^2} \propto \frac{1}{N^{2/3}} \propto \frac{1}{L^2} \qquad (1.6)$$

with *L* a length characterizing the system size. This leads to a Gaussian distribution near the mean that has the form

$$\rho(W) \propto \exp\left[-cL^2 \left(\frac{W}{\left\langle W \right\rangle} - 1 \right)^2 \right] \qquad (1.7)$$

with *c* a constant of $O(1)$. Fundamentally the power of *L* in the distribution has changed making this distribution much broader than the work distribution for systems with short-ranged correlations. This in turn suggests that in this case $\varepsilon_\Omega \propto e^{c'L^2}$, with *c'* a constant of $O(1)$. That is long-ranged correlations make ε_Ω much smaller than it is in the short –ranged case in the limit of large system size. Things actually are more complicated than indicated here. We show that the detailed form of ε_Ω is determined by the tails of the distribution and not by the Gaussian distribution near the mean. Nevertheless the above argument strongly suggest that ε_Ω in systems with long-ranged correlations is in general smaller than it is in the short-ranged case , and this will extend the usefulness of the Jarzynski fluctuation theorem.

 In this work we will consider a slab of ferromagnetic material where the extensions (linear dimension $L_\perp$) in the x-y plane will be taken to be large while the extension in the z - direction (also the direction of the external magnetic field) is a smaller length L . We will define the dimensionless area A as $A = L_\perp^2 / L^2$. This dimensionless area A can be large if $L_\perp \gg L$ but can also be of $O(1)$ when the two lengths are comparable. The comparison between L and the correlation length ξ will determine whether the system is long-ranged (long- ranged correlation implies $\xi \gg L$) or not. We will see that the transition from long-ranged to short-ranged correlations will be governed by a dimensionless parameter $L_h^2 = hL^2 / MJ$ where M is the magnetization and J is the exchange coupling between the local magnetic moments . For large values of L_h^2 (short- ranged correlations), it will be seen that the probability distribution is very sharp and centered around $h^{3/2}$. This will be

4

analogous to the short-ranged cases discussed above and one will find large fluctuations around the quantity $< \exp(-W / k_B T) >$, similar to the result shown in Eq. (1.5). For small values of L_h^2 , the distribution will become broad reducing considerably the fluctuations ε_Ω as defined in Eq. (1.4).

The primary result of this work is that

$$\varepsilon_\Omega \propto \exp(Af(L_h)) \qquad (1.8)$$

where $f(x)$ is a crossover function . For infinitesimal magnetic fields when correlations are long- ranged and $L_h << 1$, $f(L_h) \propto L_h^4$, while for larger magnetic fields , when correlations are short ranged and $L_h >> 1$, we find $f(L_h) \propto L_h^3$. In the latter case we are back to Eq. (1.5) and the usefulness of the equality in Eq. (1.1) becomes restricted to single molecules and nano scales , while in the former case we have a smaller value of ε_Ω and the usefulness of the equality can be expanded to mesoscopic and even macroscopic systems. It should be borne is mind though that for $W > 0$ the expectation value shown in Eq. (1.1) is at most unity while the root –mean- square fluctuation around the expectation value as defined in Eq. (1.4) is always greater than unity.

The paper is organized as follows. In Sec II we review the nature of the thermal fluctuations in a ferromagnet . The moments of the work fluctuations are considered in Sec III and the resulting probability distribution in Sec IV. Our results are summarized in Sec V. Details about the average work and the shape of the work probability distribution are further elucidated in an Appendix.

II A. THERMAL FLUCTUATIONS IN A FERROMAGNET

Recalling the statistical mechanics of a three- dimensional ferromagnet in an external magnetic field in the z- direction at very low temperatures , we note that the total magnetization $\vec{M}$ can be considered to be of fixed magnitude with most of its contribution coming from the large value M_0 in the z-direction. The transverse magnetization components (the Goldstone modes of the system) constitute a two- dimensional vector $\vec{m}$ with components m_1, m_2 which at these low temperatures can be considered to be small fluctuations. The components of the magnetization vector satisfy the constraint

$$m_1^2 + m_2^2 + M_0^2 = M^2 \qquad (2.1)$$

5

with M treated as a constant as explained. The free energy of the system (non-linear sigma model) can be written as [29-31]

$$F = \frac{1}{V}\int d^3r\left[\frac{J}{2}\left((\vec{\nabla} m_1)^2 + (\vec{\nabla} m_2)^2 + (\vec{\nabla} M_0)^2\right) - hM_0\right] \qquad (2.2)$$

where J is the strength of the coupling between the neighbouring transverse fluctuations and $V = L_\perp^2 L$ is the volume of the system. From Eq. (2.1), $M_0 \cong M - (m_1^2 + m_2^2)/2M$ and, hence, to quadratic order the free energy becomes

$$F = \frac{1}{V}\int d^3r\sum_{i=1}^{2}\left[\frac{J}{2}(\vec{\nabla} m_i)^2 + \frac{h}{2M} m_i^2\right] \qquad (2.3)$$

An important question is whether or not the quadratic expression given by Eq. (2.3) is sufficient for our purposes. Since we are deep in the ordered phase, the system can be characterized by a stable low- temperature renormalization group (RG) fixed point (see appendices in refs [32], [33] and [35]). In these references it is shown that the non-quadratic corrections to Eq. (2.3) are RG irrelevant at this fixed point.

The correlation function $\left\langle m_i(\vec{x})m_j(\vec{x}+\vec{r})\right\rangle$ defines the properties of the system and can be found from the corresponding correlation function in Fourier space. The dependence of the latter on the wave vector $\vec{k}$ can be easily deduced from Eq. (2.3) :

$$\left\langle m_i(\vec{k})m_j(\vec{k}')\right\rangle = \frac{k_B T}{Jk^2 + \dfrac{h}{M}}\delta(\vec{k}+\vec{k}')\delta_{ij} \qquad (2.4)$$

where T is the temperature at which the system is kept. The crossover that we mentioned earlier is evident from the above formula. For $h \to 0$ (infinitesimal magnetic field), the correlation function is proportional to k^{-2}, which in co-ordinate space leads to a long-ranged r^{-1} behaviour. For finite values of the magnetic field h, the correlation function falls off exponentially in space with a correlation length equal to $(MJ/h)^{1/2}$.

The system is now taken to be of finite extent in the z-direction by considering the extension in that direction to be of length L which forces the magnetization fluctuations to be described by a Fourier series in the z- direction while the Fourier transform can still be used in the horizontal plane with area $L_\perp^2$. The dimensionless area is $A = L_\perp^2 / L^2$. In the z-direction large and small lengths are defined with respect to the correlation length . The Fourier decomposition of the magnetization fluctuation (m_1 or m_2) which we will simply denote by $m(\vec{r})$, unless it is essential to differentiate between the components, can be written as

6

$$m(\vec{r}) = \int \frac{d^2 p}{(2\pi)^2} \sum_{n=1}^{\infty} \frac{2}{L} e^{i\vec{p}.\vec{r}_\perp} \sin \frac{n\pi z}{L} m(\vec{p}, n) \qquad (2.5)$$

where $\vec{p}$ is a two- dimensional vector in the horizontal ($x - y$) plane and we have taken the magnetization fluctuations to vanish at the surfaces $z = 0$ and $z = L$. The correlation function of Eq. (2.4) now becomes

$$\langle m(\vec{p}, n) m(\vec{q}, n') \rangle = \frac{k_B T L / 2}{J(p^2 + \frac{n^2 \pi^2}{L^2}) + \frac{h}{M}} \delta(\vec{p} + \vec{q}) \delta_{nn'} \qquad (2.6)$$

IIB. WORK DONE IN FERROMAGNETS AS THE MAGNETIC FIELD IS CHANGED

We now consider the work done as the magnetic field in which a ferromagnet is placed is changed from an initial $h = 0$ to a final value 'h'. It will be defined as

$$W_E = -\int_0^h dh' M_0 \cong -Mh + \int_0^h dh'(m_1^2 + m_2^2) / 2M \qquad (2.7)$$

We need to comment on the definition of the work W above for the magnetic system. In analogy with the work done for the expanding gas discussed in section I, one would have expected the differential work for the magnetic system to be $dW = hdM$. However, it is simpler to do calculations with Eq. (2.7). To this end we define a Gibbs free-energy $G = U - TS - hM$, so that for an isothermal process, the dissipative entropy satisfies $TdS - dQ = dW_E - dG$. Here dW_E is an 'enthalpy' work, U is the internal energy and dQ is the heat energy. The Jarzynski equality (Eq. (1.1)), when the magnetic work is W_E of Eq. (2.7), becomes $\langle e^{-W_E/kT} \rangle = e^{-\Delta G/kT}$. The Gibbs free energy appears on the right- hand side of Eq. (1.1) when the work done is the 'enthalpy' work defined above. In what follows we drop the subscript E. Returning to Eq. (2.7), the first term is a constant and the second, when averaged over the thermal fluctuations, is the local work fluctuation $W_{fl}(\vec{r})$ to the lowest order. We define

$$W_{fl}(\vec{r}) = \int dh' < (m_1(\vec{r})^2 + m_2(\vec{r})^2) > /2M \qquad (2.8)$$

where the angular brackets above refer to an average over the thermal fluctuations. We are ignoring the higher- order terms arising from the binomial expansion of $(M^2 - m_1^2 - m_2^2)^{1/2}$ in Eq. (2.7). Our interest is in the probability distribution for $\langle W_{fl,T} \rangle$, where the subscript T indicates that we are considering the total magnetic work in the volume V, defined as

$$\langle W_{fl,T} \rangle = \int_0^h dh' \int d^3 r \langle m_1(\vec{r})^2 + m_2(\vec{r})^2 \rangle / 2M \qquad (2.9)$$

7

The probability distribution will be accessed by calculating the various moments . The n-th moment will be defined as

$$\left\langle W_{fl,T}^{n} \right\rangle = \left(\frac{1}{2M} \right)^{n} \int_{0}^{h} dh_{1} .. \int_{0}^{h} dh_{n} \int d^{3}r_{1} ... \int d^{3}r_{n} \left\langle (m_{1}^{2}(\vec{r}_{1}) + m_{2}^{2}(\vec{r}_{1})) ...... (m_{1}^{2}(\vec{r}_{n}) + m_{2}^{2}(\vec{r}_{n})) \right\rangle \qquad (2.10)$$

For n=1, the average value $\left\langle W_{fl,T} \right\rangle$ is found on calculation to be much smaller than Mh, which justifies ignoring the higher order terms coming from the binomial expansion of $\sqrt{M^{2} - m_{1}^{2} - m_{2}^{2}}$ in Eq.(2.7). Since fluctuations in directions '1' and '2' are not correlated, Eq.(2.10) can be written as

$$\left\langle W_{fl,T}^{n} \right\rangle = 2 \left(\frac{1}{2M} \right)^{n} \int_{0}^{h} dh_{1} .. \int_{0}^{h} dh_{n} \int d^{3}r_{1} .. \int d^{3}r_{n} \left\langle m^{2}(\vec{r}_{1}) m^{2}(\vec{r}_{2}) .... m^{2}(\vec{r}_{n}) \right\rangle \qquad (2.11)$$

In the next section we shall calculate the moments (actually only the cumulants) and in Section IV we shall obtain the asymptotic form of the probability distribution.

III. MOMENTS OF THE WORK FLUCTUATIONS

We start with the first moment which is the average of the work fluctuations (n =1 in Eq. (2.11)) and find

$$\left\langle W_{fl,T} \right\rangle = \frac{1}{MV} \int_{0}^{h} dh' \int d^{3}r \left\langle m(\vec{r})^{2} \right\rangle$$

$$= \frac{1}{M} \int_{0}^{h} dh' \int d^{3}r \sum_{n_{1},n_{2}} \frac{4}{L^{2}} \sin \frac{n_{1}\pi z}{L} \sin \frac{n_{2}\pi z}{L} \int \frac{d^{2}p}{(2\pi)^{2}} \int \frac{d^{2}q}{(2\pi)^{2}} e^{i(\vec{p}+\vec{q}).\vec{r}} \left\langle m(\vec{p}, n_{1}) m(\vec{q}, n_{2}) \right\rangle$$

$$= \frac{2}{ML^{2}} \int_{0}^{h} dh' \int d^{3}r \sum_{n=1}^{\infty} \int \frac{d^{2}p}{(2\pi)^{2}} \sin^{2} \frac{n\pi z}{L} \frac{k_{B}TL}{J(p^{2} + \frac{n^{2}\pi^{2}}{L^{2}}) + \frac{h'}{M}}$$

$$= \frac{L_{\perp}^{2}}{L^{2}} \frac{L^{2}}{MJ} \int_{0}^{h} dh' \sum_{n=1}^{\infty} \int \frac{d^{2}(pL)}{(2\pi)^{2}} \frac{k_{B}T}{p^{2}L^{2} + n^{2}\pi^{2} + \frac{h'L^{2}}{MJ}} \qquad (3.1)$$

From now on we label the dimensionless wave number pL as q and define

$$\gamma^{2} = p^{2}L^{2} + \frac{h'L^{2}}{MJ} = q^{2} + \frac{h'L^{2}}{MJ}, \qquad (3.2)$$

8

After performing the summation over 'n' and introducing an ultraviolet cut-off at $p = \Lambda \approx a^{-1}$ (here 'a' is the lattice spacing), we write Eq. (3.1) as

$$\left\langle W_{\text{fl,T}} \right\rangle = k_B T \frac{L_\perp^2}{L^2} \frac{L^2}{MJ} \int_0^h dh' \int_0^{\Lambda^2 L^2} \frac{d(q^2)}{4\pi} \frac{1}{2\gamma} (\coth \gamma - \frac{1}{\gamma})$$

$$= \frac{1}{4\pi} k_B T A L_h^2 \int_0^1 dx \int_{\left(\frac{hL^2}{MJ}\right)^{1/2}}^{\Lambda L} d\gamma (\coth \gamma - \frac{1}{\gamma}) \qquad (3.3)$$

where as defined before $A = L_\perp^2 / L^2$ and $L_h^2 = hL^2 / MJ$. Evaluating the integral in Eq. (3.3), we now have

$$\left\langle W_{\text{fl,T}} \right\rangle = k_B T \frac{A L_h^2}{4\pi} \int_0^1 dx \left[\ln \frac{\sinh \Lambda L}{\Lambda L} - \ln \frac{\sinh \sqrt{x L_h^2}}{\sqrt{x L_h^2}} \right]$$

$$= k_B T \frac{A L_h^2}{4\pi} \left[\Lambda L - \ln(2\Lambda L) - \int_0^1 dx \ln \frac{\sinh \sqrt{x L_h^2}}{\sqrt{x L_h^2}} \right] \qquad (3.4)$$

The first two terms in the square bracket above, which are cut-off dependent, arise from the divergence of the integral. We note that this divergence stems from the large wave- number dependence of the integrand and thus corresponds to the short- ranged correlations. It should be noted that the average work is proportional to the volume of the system because of this divergence and is very different from the higher moments. We will return to this issue in the Appendix. The crossover effect is present in the finite part represented by the integral which is zero for $L_h^2 \ll 1$, while for $L_h^2 \gg 1$ the dominant term comes from the growing exponential inside the log and it is easily seen to be $2L_h / 3$. We note that the high-momentum divergence that we see in the first moment will not occur in the higher moments. It should also be noted that in the high- field limit the dependence L_h^3 that has emerged will remain throughout regardless of the degree of the moment.

We now turn to the second moment (actually the cumulant) which will be the prototype for all other cumulants since it will not have the divergence that plagues the first moment. We will do this in some detail and then generalize to the higher moments. Setting n =2 in Eq. (2.11), we consider

$$\left\langle W_{\text{fl,T}}^2 \right\rangle = 2 \left(\frac{1}{2M} \right)^2 \int_0^h dh'' \int_0^h dh' \int d^3 r_1 \int d^3 r_2 \left\langle m_{h'}^2(\vec{r}_1) m_{h''}^2(\vec{r}_2) \right\rangle \qquad (3.5)$$

9

The subscripts associated with the two different $m^2(\vec{r})$ above make it clear that the process $h'=0$ to $h'=h$ will in general be different for the different $m^2(\vec{r})$. A Gaussian distribution for the magnetization fluctuations will imply a factorization of the correlation function shown above and we have for the second-order cumulant

$$\left\langle W_{fl,T}^2 \right\rangle_{cum} = 2\times 2\left(\frac{1}{2M}\right)^2 \int_0^h dh'' \int_0^h dh' \int d^3 r_1 \int d^3 r_2 \left\langle m_{h'}(r_1)m_{h''}(r_2)\right\rangle^2 \qquad (3.6)$$

In the above we have shown the first factor of 2 separately as it is a combinatorial factor that will change with n. To find the equilibrium equal-time correlation that we need in Eq. (3.6), it is simplest to do the calculation in Fourier space and evaluate $\left\langle m_h(\vec{k},t)m_{h'}(-\vec{k},t)\right\rangle$.

We begin by writing the evolution equation for the Fourier transform $m_h(\vec{k},t)$. The dynamics is the usual Langevin dynamics with the noise having the appropriate statistical property to give the equilibrium correlation function of Eq. (2.4). Explicitly,

$$\dot{m}_h(\vec{k},t) = -\Gamma(Jk^2 + \frac{h}{M})m(\vec{k},t) + \varsigma(\vec{k},t) \qquad (3.7)$$

where $\varsigma(\vec{k},t)$ is the delta-correlated Gaussian white noise with the correlation function

$$\left\langle \varsigma(\vec{k}_1,t_1)\varsigma(\vec{k}_2,t_2)\right\rangle = 2\Gamma k_B TV(Jk_1^2 + \frac{h}{M})\delta(\vec{k}_1 + \vec{k}_2)\delta(t_1 - t_2) \qquad (3.8)$$

After integrating

$$m_h(\vec{k},t) = \int dt' \varsigma(\vec{k},t')e^{-\Gamma(Jk^2 + \frac{h}{M})(t-t')} + m_h(\vec{k},0)e^{-\Gamma(Jk^2 + \frac{h}{M})t} \qquad (3.9)$$

The equilibrium correlation function is obtained as the equal-time correlation function at long times and is seen to be

$$\left\langle m_h(\vec{k})m_{h'}(\vec{k}')\right\rangle = \frac{k_B TV}{Jk^2 + \frac{h+h'}{2M}}\delta(\vec{k} + \vec{k}'), \qquad (3.10)$$

giving the standard equilibrium correlation function of Eq. (2.4) when $h=h'$.

Returning to Eq. (3.6), we write

$$\left\langle m_{h'}(\vec{r}_1)m_{h''}(\vec{r}_2)\right\rangle = \sum_{n_1=1}^{\infty} \frac{4}{L^2}\sin\frac{n_1\pi z}{L}\sum_{n_2=1}^{\infty}\sin\frac{n_2\pi z}{L}\int\frac{d^2 p}{(2\pi)^2}\int\frac{d^2 q}{(2\pi)^2}e^{i\vec{p}.\vec{r}_{1\perp}+i\vec{q}.r_{2\perp}}\left\langle m_{h'}(\vec{p},n_1)m_{h''}(\vec{q},n_2)\right\rangle$$

$$= \sum_{n=1}^{\infty} \frac{2}{L} \sin \frac{n\pi z_1}{L} \sin \frac{n\pi z_2}{L} \int \frac{d^2 p}{(2\pi)^2} e^{i\vec{p}\cdot(\vec{r}_{1\perp}-\vec{r}_{2\perp})} \frac{k_B T}{J(p^2 + \frac{n^2\pi^2}{L^2}) + \frac{h''+h'}{2M}} \qquad (3.11)$$

To evaluate the integral in Eq. (3.6), we need another such factor with n replaced by n' and $\vec{p}$ replaced by $\vec{q}$. The integral over z_1 will then yield a factor of $L\delta_{nn'}/2$ and the integration over z_2 gives $L/2$. Similarly, integration over $r_{1\perp}$ yields $\delta(\vec{p}+\vec{q})$ and the integration over $r_{2\perp}$ gives $L_{\perp}^2$. This allows us to write Eq. (3.6) as

$$\left\langle W_{\mathrm{fl,T}}^2 \right\rangle_{\mathrm{cum}} = 2 \times 2 \left(\frac{1}{2M} \right)^2 \int_0^h dh'' \int_0^h dh' \sum_{n=1}^{\infty} \int \frac{d^2 p}{(2\pi)^2} \left[\frac{k_B T}{J(p^2 + \frac{n^2\pi^2}{L^2}) + \frac{h''+h'}{2M}} \right]^2 \qquad (3.12)$$

Carrying out the rescalings described in the calculation of $\left\langle W_{\mathrm{fl}} \right\rangle$ above , we write Eq. (3.12) as

$$\left\langle W_{\mathrm{fl,T}}^2 \right\rangle_{\mathrm{cum}} = 2 \times \frac{1}{2}(2k_B T L_h^2)^2 A \int_0^1 dx'' \int_0^1 dx' \sum_{n=1}^{\infty} \int \frac{d(q^2)}{4\pi} \left(\frac{1}{q^2 + n^2\pi^2 + L_h^2 \frac{x''+x'}{2}} \right)^2$$

$$= \frac{1}{4\pi}(2k_B T L_h^2)^2 A \int_0^1 dx'' \int_0^1 dx' \sum_{n=1}^{\infty} \frac{1}{n^2\pi^2 + L_h^2 \frac{x''+x'}{2}}$$

$$= \frac{1}{4\pi}(2k_B T L_h^2)^2 A \int_0^1 dx'' \int_0^1 dx' \frac{1}{2\gamma'}\left(\coth\gamma' - \frac{1}{\gamma'}\right) \qquad (3.13)$$

The γ' in the last line of Eq. (3.13) is defined as $\gamma'^2 = L_h^2(x''+x')/2$. We now examine this formula in the two limiting cases of $L_h^2 \ll 1$ and $L_h^2 \gg 1$.

i) $L_h^2 \ll 1$: In this limit $\gamma' \to 0$ and the first non- vanishing term of the integrand in Eq. (3.13) is seen to be 1/3 and, hence,

$$\left\langle W_{\mathrm{fl,T}}^2 \right\rangle_{\mathrm{cum}} = \frac{1}{4\pi}(2k_B T L_h^2)^2 A \left[\frac{1}{6} + O(L_h^4) \right] \qquad (3.14)$$

ii) $L_h^2 \gg 1$: In this limit $\gamma' \to \infty$ and the leading contribution comes from the region where $\coth\gamma' = 1$ and we find

11

$$\left\langle W_{\text{fl,T}}^2 \right\rangle_{\text{cum}} = \frac{1}{4\pi}(2k_BT)^2 \, AL_h^3 \int_0^1 dx" \int_0^1 dx' \frac{1}{2(x"+x')^{1/2}} = \frac{1}{4\pi}(2k_BT)^2 \, AL_h^3 \frac{4}{3}(\sqrt{2}-1) \quad (3.15)$$

In this limit the next term is of $O(L_h^2)$.

We note that while the finite part of $\left\langle W_{\text{fl,T}} \right\rangle$ would be $O(L_h^2)$ [it is accidental that the coefficient is zero] in the small L_h limit, the corresponding dependence of $\left\langle W_{\text{fl,T}}^2 \right\rangle_{\text{cum}}$ is $O(L_h^4)$ (Eq. (3.14)) and for $\left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}}$ it will be $O(L_h^{2n})$ as we will argue below. However, for very large values of L_h , the dependence of $\left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}}$ will always be L_h^3 regardless of n . For any 'n',we can write

$$\left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}} = 2^{n-1}\,(n-1)! \times 2\left(\frac{1}{2M}\right)^n \int_0^h dh_1 ... \int_0^h dh_n \int d^3 r_1 ... \int d^3 r_n \left\langle m_{h_1}(\vec{r}_1) m_{h_2}(\vec{r}_2) \right\rangle ....$$

$$..........\left\langle m_{h_n}(\vec{r}_n) m_{h_1}(\vec{r}_1) \right\rangle \qquad (3.16)$$

Inserting the appropriate Fourier expansions and performing the spatial integrations , we obtain

$$\left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}} = \frac{1}{4\pi}(n-1)!\,A(2k_BTL_h^2)^n \int_0^1 dx_1 ... \int_0^1 dx_n \sum_{s=1}^{\infty} \int_0^{\infty} d(q^2) \frac{1}{q^2 + s^2\pi^2 + L_h^2 \frac{x_1 + x_2}{2}} \frac{1}{q^2 + s^2\pi^2 + L_h^2 \frac{x_2 + x_3}{2}} ....$$

$$..... \frac{1}{q^2 + s^2\pi^2 + L_h^2 \frac{x_n + x_1}{2}} \qquad (3.17)$$

In the limit of infinitesimal fields , $L_h \to 0$, we find ($n \geq 2$)

$$\left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}} (L_h \to 0) = \frac{\pi}{4}(n-2)!\,A\left(\frac{2k_BT}{\pi^2}\right)^n L_h^{2n}\varsigma(2n-2) \qquad (3.18)$$

where $\varsigma(x)$ is the usual Riemann zeta function. On the other hand, in the limit of large L_h , the scaling behaviour can be inferred by noting that the integrand in Eq. (3.17) can be written as partial fractions where each term has the form $(q^2 + s^2\pi^2 + L_h^2 \frac{x_i + x_j}{2})^{-1}$ multiplied by $n-1$ terms of the form $(L_h^2 \frac{x_l - x_k}{2})^{-1}$. These n -1 terms reduce the power of L_h^2 in the prefactor of Eq. (3.17) to unity and the remaining L_h dependence comes from the

12

sum and integral $\sum_{s=1}^{\infty} \int d(q^2)[q^2 + s^2\pi^2 + L_h^2 \frac{x_i + x_j}{2}]^{-1}$, which after summation over 's' can

be written in the leading order for large L_h^2 as $\int d(q^2)/[q^2 + L_h^2 \frac{x_i + x_j}{2}]^{1/2}$. This integral

scales as L_h and hence for $L_h \gg 1$, the leading term of Eq. (3.17) is

$$\left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}} (L_h \to \infty) = (n-2)! A \left(\frac{2k_B T}{\pi^2} \right)^n C_n L_h^3 \qquad (3.19)$$

where C_n is a number of order unity. The dependence on h follows from the general result
that the Goldstone-mode-dominated susceptibility scales as L_h^{-1} [34,35]. We note that all
the moments are proportional to the system size $L_\perp^2 L$ as happens always in a system with
short- ranged correlations. In sharp contrast , when one is dealing with long- ranged
correlations ($L_h \to 0$) , all the moments (except the average work) are proportional to
$L_\perp^2 / L^2$ as shown in Eq. (3.18).

What kind of a crossover function are we finding ? To answer this it is easiest to examine
higher- order terms in Eq. (3.17) as L_h is increased. Expanding the denominator of each
factor in the integrand , it is clear that the series is going to be alternating and the first
correction is

$$\left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}} = \left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}} (L_h \to 0) \left[1 - \frac{n-1}{2} \frac{\zeta(2n-4)}{\zeta(2n-2)} L_h^2 + O(L_h^4) \right] \qquad (3.20)$$

An interpolation which guarantees that coefficients come out correctly for large n (in
practice $n > 4$, when the zeta functions are effectively unity) is

$$\left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}} = \left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}} (L_h \to 0) \left[1 + L_h^2 \right]^{3/2} / \left[1 + \frac{L_h^2}{2} \right]^n \qquad (3.21)$$

For the special case of $n=2$, we introduce the crossover function $f(L_h)$ which gets the
coefficients correctly in both the small and large L_h limits and write

$$\left\langle W_{\text{fl,T}}^2 \right\rangle_{\text{cum}} = (k_B T)^2 A f(L_h) \qquad (3.22)$$

An explicit interpolation form for $f(L_h)$ is

$$f(L_h) = \frac{L_h^4}{6\pi \left(1 + \frac{L_h}{8(\sqrt{2} - 1)} \right)} \qquad (3.23)$$

13

which as we will see below is the function that will describe the crossover of the fluctuation ε_Ω, introduced in Eq. (1.8). Having obtained the moments we now construct the probability distribution in the next section.

IV   THE PROBABILITY DISTRIBUTION

We find the work probability distribution $\rho(W)$ by constructing a cumulant generating function $K(t)$ defined by

$$K(t) = \sum_{n=1}^{\infty} \frac{\left\langle W_{\text{fl,T}}^n \right\rangle_{\text{cum}} t^n}{n!} \qquad (4.1)$$

In this sum the terms $n \geq 2$ are determined from Eqs. (3.18) and (3.21), while for $n = 1$, we need the leading term (proportional to the size of the system) of Eq. (3.4). It should be noted that though for large n, the cumulants $\left\langle W_{\text{fl,T}}^n \right\rangle$ are very large, the series for $K(t)$ is always convergent due to the factor of $n!$ in the denominator of the right hand side of Eq.(4.1). Indeed, the precise condition [36] for the moments to determine the distribution function is that the moment generating function $K(t)$ above has a finite radius of convergence. The probability distribution function is then obtained by takingthe inverse Laplace transform

$$\rho(W) = \int_0^{\infty} e^{-Wt + K(t)} dt \qquad (4.2)$$

The integral in Eq. (4.2) will be evaluated by the method of steepest descent, since the evaluations of $K(t)$ or $\rho(W)$ can not be done exactly. In the sum shown in Eq. (4.1), the first two terms determine the Gaussian approximation to the distribution function. The higher- order terms ($n > 2$) are responsible for the tails of the distribution function. To get the tails accurately, we need the larger values of n and as a result $n - 1$ can be replaced by n and the zeta functions set to unity ($\varsigma(4) = \pi^4 / 90$ is already very close to unity). For $W > 0$, we have

$$K(t) = \frac{\pi}{4} A (1 + L_h^2)^{3/2} \sum_n (k_B T)^n \left(\frac{2 L_h^2}{\pi^2 (1 + \frac{L_h^2}{2})} \right)^n \frac{t^n}{n^2}$$

$$= a \sum_n \frac{(bt)^n}{n^2} \qquad (4.3)$$

with

14

$$a = \pi A (1 + L_h^2)^{3/2} / 4, b = k_B T \frac{4 L_h^2}{\pi^2 (2 + L_h^2)} \qquad (4.4)$$

It should be noted that the $K(t)$ of Eq. (4.3) gives an average value of W that is $\langle W \rangle = ab$, different from the result obtained in Eq. (3.4). This will be important when we discuss the Gaussian approximation below.

From Eq. (4.3),we see that

$$K'(t) \equiv \frac{dK}{dt} = \frac{a}{t} \ln \frac{1}{(1 - bt)} \qquad (4.5)$$

The saddle- point method for obtaining $\rho(W)$ requires finding the saddle point t_0 where

$$W = K'(t_0) \qquad (4.6)$$

and writing

$$\rho(W) = C e^{-W t_0 + K(t_0)} \qquad (4.7)$$

where C is a constant obtained from normalization . We need to find t_0 from the requirement

$$W = -\frac{ab}{bt_0} \ln(1 - bt_0) = -\frac{ab}{x_0} \ln(1 - x_0) \qquad (4.8)$$

where $x = bt$. The scale of W is set by ab . We need to emphasize again that this scale is not necessarily the average value of W_{fl}. For small values of L_h^2 the dimensionless scale ($ab / k_B T$) is set by $A L_h^2 / 2\pi$, which scales as hV / L^3. For large values of L_h^2, the scale is set by $A L_h^3 / \pi$ which scales as $h^{3/2} V$. This scale , like the average work in Eq. (3.4), is proportional to the system size . In this case , things are reasonably clear- all the action is centred around the scale 'ab'. Regions close to this scale and far away from it can be explored by expanding the right- hand side of Eq. (4.8) in a Taylor series and determining x_0 . The probability distribution follows from Eq. (4.7). We represent the scale factor by the crossover form (see Eq. (4.4))

$$\frac{ab}{k_B T} = \frac{A}{\pi} \frac{\left(1 + L_h^2\right)^{3/2}}{\left(1 + \dfrac{2}{L_h^2}\right)} = \frac{A}{\pi} S(L_h) \qquad (4.9)$$

Writing the dimensionless work in units of $k_B T$ in Eq. (4.8) , we find

$$\pi W = -AS(L_h)\frac{\ln(1-x_0)}{x_0} \qquad (4.10)$$

For both $L_h \gg 1$ and $L_h \ll 1$, large values of W correspond to $x_0 \cong 1$. Writing $x_0 = 1 - \varepsilon$, we see that $\varepsilon \cong \exp[-\pi W / AS]$. The integral of Eq. (4.5) can be written as

$$K(t) = -a\left[\ln x \ln(1-x) + \int \frac{\ln x}{1-x}dx\right] + K_0 \qquad (4.11)$$

where K_0 is a constant. All terms in $K(1-\varepsilon)$ are at least $O(\varepsilon)$ and hence the asymptotic large W behaviour of $\rho(W)$ is $\exp(-W / b)$. The scale of W in Eq. (4.8) changes with changing L_h but the form of $\rho(W)$ is unaltered.

For $x_0 \ll 1$, one is in the vicinity of $W = AS(L_h) / \pi$, which is proportional to L_h^2 for small magnetic fields and to L_h^3 for large fields. In this range, $x_0 \cong 2\left(\frac{W\pi}{AS(L_h)} - 1\right)$ and the distribution, in this approximation, is a Gaussian centered at $AS(L_h)$. This, as expected , is different from the exact Gaussian obtained by keeping the $n=1$ and $n=2$ terms in Eq. (4.1).

The limit of very small W needs to be handled carefully and we note that the only way one can achieve small values of $-\frac{\ln(1-x_0)}{x_0}$ is by going to large negative values of x_0 so that Eq. (4.8) becomes

$$\pi W = AS(L_h)\frac{\ln(1+|x_0|)}{|x_0|} \qquad (4.12)$$

Inverting , we obtain

$$|x_0| \cong AS(L_h)\frac{\ln(AS(L_h)\pi^{-1}W^{-1})}{\pi W} \qquad (4.13)$$

Integration of Eq. (4.5) for large negative values of t , yields

$$K(t) = -\frac{a}{2}\left(\ln|x|\right)^2 \qquad (4.14)$$

For $W \to 0$, we use Eq. (4.13) to calculate the exponent $-Wt_0 + K(t_0)$ (we are not showing any prefactors since they will not be relevant) and get

$$\rho(W \to 0) \propto e^{-\frac{a}{2}\left(\ln\frac{AS(L_h)}{\pi W}\right)^2 + a\ln\left(\frac{AS(L_h)}{\pi W}\right)} \qquad (4.15)$$

16

For W in the vicinity of ab, we have the Gaussian form (note that this Gaussian form stems from the approximate form of $K(t)$ in Eq. (4.3)) given by

$$\rho(W) \propto \exp\left(-\frac{(W-ab)^2}{ab^2}\right) \qquad (4.16)$$

Using the $n=1$ and $n=2$ terms of Eq. (4.1), we obtain the more general Gaussian form,

$$\rho(W) \propto e^{-\frac{(W-\langle\langle W\rangle\rangle)^2}{2\langle W_{fl}^2\rangle}} = e^{-\frac{<W>^2}{2<W_{fl}^2>}\left(\frac{W}{<W>}-1\right)^2} = e^{-\frac{<W>^2}{2Af(L_h)}\left(\frac{W}{<W>}-1\right)^2} \qquad (4.16a)$$

For $W \gg ab$, we find

$$\rho(W) \propto \exp(-W/b) \qquad (4.17)$$

Eqs. (4.15)-(4.17) constitute the basis for the primary result of this paper quoted in Eq. (1.8).

Specifically, for the work distribution in the case of long- ranged correlation ($L_h < 1$) we obtain :

i) $\quad W \ll AL_h^2/2\pi, \quad \rho(W) \propto \exp\left[-\frac{\pi A}{8}\left(\ln\frac{AL_h^2}{2\pi W}\right)^2 + \frac{\pi A}{4}\ln\left(\frac{AL_h^2}{2\pi W}\right)\right] \qquad (4.18a)$

ii) $\quad W \approx <W> = AL_h^2/2\pi, \quad \rho(W) \propto \exp\left[-\frac{\pi A(\frac{2\pi W}{AL_h^2}-1)^2}{4}\right] \qquad (4.18b)$

iii) $\quad W \gg AL_h^2/2\pi \quad \rho(W) \propto \exp\left(-\pi^2 W/2L_h^2\right) \qquad (4.18c)$

The results i) and iii) are identical to what was reported in Kirkpatrick *et al.* [23]. For ii), the Gaussian form shown in Eq. (4.18b) is based on Eq. (4.16) while the form shown in reference [28] was based on Eq. (4.16a) . The difference does not matter in establishing the primary point , which is the scale of the probability distribution. The difference between Eqs. (4.16a) and (4.18b) will be commented upon in the Appendix.

The corresponding results ρ_{SR} for the short- ranged correlations, as follows from Crooks and Jarzynski [12], are

$$\rho_{SR}(W \to 0) \propto e^{-\frac{3N}{2}\ln\left(\frac{<W>}{W}\right)} \qquad (4.19a)$$

$$\rho_{SR}(W \approx <W>) \propto e^{-\frac{3N}{2}(\frac{W}{<W>}-1)^2} \qquad (4.19b)$$

$$\rho_{SR}\,(W>>< W >) \propto e^{-\frac{3NW}{2<W>}} \qquad\qquad (4.19c)$$

The important difference between these short- ranged probability distributions and the long- ranged ones of Eqs. (4.18a) and (4.18b) lies in the scale of the distribution. In the long-ranged case it is set by A which is proportional to V/L^3 and for the short- ranged case it is set by N which is equal to V/a^3 where a is the lattice spacing . For $W >> \langle W \rangle$, the distributions are similar. In the evaluation of the moments of Ω, it is the $W \le \langle W \rangle$ parts which matter and hence the suppression of the probability distribution for the short- ranged correlation leads to a larger ε_Ω .

The fact that the long-ranged correlations lead to a much larger tail than the short-ranged ones is best seen through a numerical example. We consider a specific case of a typical protein for which $L_\perp = 10a$ and $L = 4a$. This makes A= 6.25. We consider two values of W such that $W << \langle W \rangle$ and $W \approx \langle W \rangle$, to make our point. The value of N for this case can be found from $Na^3 = V = L_\perp^2 L$ and is seen to be 400. For $W = \langle W \rangle /100$, which is far from the peak, $\rho(W)/\rho_{SR}(W) \approx e^{1317}$, showing that in the tail of the probability distribution, for the long-ranged case the probability is enormous when compared to the short-ranged case. For $W = \langle W \rangle /2$,which is close to the peak, this ratio is e^{149} and the long-ranged case probability is still much greater than the short-ranged case. This makes the point that the systems with long-ranged correlations have a much broader probability distribution for the work done.

We now turn to the calculation of the fluctuation ε_Ω defined in Eq. (1.5). We use the distribution shown in Eq. (4.16a) for this purpose. Carrying out the Gaussian integrals,

$$\left\langle e^{-nW} \right\rangle = e^{n^2 Af(L_h)/2} \qquad (4.20)$$

Consequently,

$$\varepsilon_\Omega = \frac{\left\langle \Omega^2 \right\rangle}{\left\langle \Omega \right\rangle^2} - 1 \cong \frac{\left\langle \Omega^2 \right\rangle}{\left\langle \Omega \right\rangle^2} = e^{Af(L_h)} \qquad (4.21)$$

with $f(x)$ defined in Eqs. (3.22) and (3.23). We have thus established Eq. (1.6) which was stated to be the primary result of the paper. If we take the example cited above ($A = 6.25$, $N = 400$) then for the long-ranged case ($L_h << 1$), $\varepsilon_\Omega \approx e^{2L_h^4/3\pi}$ which can be quite close to unity for $L_h << 1$. For the short –ranged case, using Eq. (1.5) with $N = 400$ and assuming small α , we find $e^{600\alpha^2}$ which is astronomical unless α is very small . The root-mean-square fluctuation about the average of Ω is $e^{300\alpha^2}$ much greater than $<\Omega>$ itself which is $e^{-600\alpha^2}$. If one reduces the number of particles still further (in terms of system size, considering

18

nano scales and beyond) one can make the root-mean- square fluctuation smaller for short-ranged fluctuations but it will still be greater than the average value.

In order to provide numbers to be able to see the short-ranged to long-ranged crossover in an actual experimental set-up, it is necessary to ask the question how small must the external magnetic field be to allow us to use the long-ranged approximation , $L_h^2 \ll 1$. For this we need an estimate for L, which can be taken to be the typical size of ferromagnetic domains. Free- energy considerations restrict the domain size to 10^{-7} m to 10^{-8} m. To be definite, we take 10^{-8} m as our L. If we denote a microscopic length by l, then $L = 100l$. Writing $L_h^2 = hL^2 / MJ = L^2 / L_H^2$, we have $L_H^2 = MJ / h = M^2 J / Mh = k_B Tl^2 / \mu_B h$, where in the last step we have replaced the magnetization in the denominator by the Bohr magneton μ_B and estimated JM^2 by appealing to Eq. (2.3) and writing $k_B T$ for the free energy. At $T = 100$ K , $k_B T \approx 10^{-21}$ Joules and with h in teslas , $\mu_B \approx 10^{-23}$ Joules/tesla. We thus estimate $L_H^2 \approx 100l^2 / h$ and $L_h^2 \approx 100h$, so that the condition $L_h^2 < 1$ implies $h < 10^{-2}$ teslas which is 10^2 Gauss.

It should be noted that the kind of experiments used to test the Jarzynski equality have been primarily single- molecule experiments ($e.g.$, Refs. [37-39]) where the probability distribution has been used to determine the free- energy difference between initial and final states [$e.g.$, the folded and unfolded states of a DNA hairpin] or the energy fluctuations in a single harmonic oscillator driven out of equilibrium by an external torque. An example of the latter is the thermal rheometer [40-43] which is a torsion pendulum whose minute angular displacements are measured by a highly sensitive interferometer. The pendulum is driven out of equilibrium by an external torque which amounts to a few pico newton- metres. The thermal fluctuations amount to a root- mean- square angular displacement of a few nano radians. This gives an idea of the scale of the system. The torsion pendulum operates in the linear regime, the probability distributions are Gaussian and the test of the fluctuations dissipation theorems depends only on the width of the distribution. What we are pointing out is that even for a mesoscopic system (the example given above) the Jarzynski equality will be difficult to test because of the large fluctuations, but if the fluctuations are long-ranged then even for a macroscopically large system ($N \approx 10^{23}$), the fluctuation ε_Ω can be reduced to be of $O(1)$.

The result for ε_Ω shown in Eq. (4.21) is not a consequence of the Gaussian form for the probability distribution. We will get a similar result if we use the low-W form of the distribution Eq. (4.18a). We are interested in calculating $\langle \Omega^n \rangle$ and we will use the probability distribution of Eq. (4.18a) to the leading order only. We need

$$\langle \Omega^n \rangle = \int \rho(W)\Omega^n \, dW \, / \int \rho(W) \, dW$$

$$= \int \rho(W)e^{-nW} \, dW \, / \int \rho(W) \, dW \qquad (4.22)$$

The calculation will entail ignoring all non-exponential pre-factors and hence the normalizing integral in the denominator will contribute unity. The numerator will be calculated in the saddle- point approximation described before. We need the integral

$$I_n = \int_0^\infty \exp\{-\frac{\pi A}{8}\left[\left(\ln\frac{AL_h^2}{2\pi W}\right)^2 + \frac{8nW}{\pi A}\right]\}dW = \int_0^\infty \exp\left[-\frac{\pi A}{8}g_n(W)\right]dW \quad (4.23)$$

In terms of the variable $x = 2\pi W / AL_h^2$, the function $g(W)$ can be written as

$$g_n(x) = \left(\ln\frac{1}{x}\right)^2 + n\frac{4L_h^2}{\pi^2}x = \left(\ln\frac{1}{x}\right)^2 + n\mu x \qquad (4.24)$$

where $\mu = 4L_h^2 / \pi^2 \ll 1$ in the infinitesimal magnetic- field range where $L_h^2 \ll 1$. For very small values of μ, the above function has a minimum very near $x = 1$ and this dominates the integrand in Eq. (4.24). The minimum is at $x_m = 1 - \delta$, where to the lowest order in μ, it is seen that $\delta = \mu/2$. Up to $O(\mu^2)$, the minimum value of $g_n(x)$ is found as

$$g_n(x_m) = n\mu - \frac{n^2\mu^2}{4} \qquad (4.25)$$

Keeping only the exponential terms, we have in this approximation

$$I_n \cong \exp\left[-\frac{\pi A}{8}(n\mu - \frac{n^2\mu^2}{4})\right] \qquad (4.26)$$

Thus we have,

$$\varepsilon_\Omega = \frac{\langle \Omega^2 \rangle}{\langle \Omega \rangle^2} - 1 = \frac{I_2}{I_1^2} - 1 \cong \exp\left[\frac{\pi A}{16}\mu^2\right] = \exp\left[A\frac{L_h^4}{\pi^3}\right] \qquad (4.27)$$

We have obtained for $L_h \ll 1$, the same result as before with a different numerical factor.

Turning to the other extreme, where $L_h^2 \gg 1$ and the energy scale as well as the scale of the distribution for small W is set by the system size , we note that

i)   For $W \ll AL_h^3$: $\rho(W) \propto \exp-\frac{\pi AL_h^3}{8}\left[\ln\frac{AL_h^3}{\pi W}\right]^2 \qquad (4.28a)$

20

ii)   For $W \cong AL_h^3$ $\rho(W) \cong \exp\left[-\dfrac{\left(W - 4\pi AL_h^3\right)^2}{16\pi AL_h^3}\right]$ (4.28b)

iii)   For $W >> AL_h^3$ $\rho(W) \cong \exp\left(-\pi^2 W / 4\right)$ (4.28c)

In the range of large magnetic field (short- ranged correlations) the factor AL_h^3 is like

$h^{3/2}V$ and the probability distribution is tremendously suppressed for the relevant regions. This completes what we wanted to describe- the passage of the work probability distribution from a broad distribution for long- ranged correlations to a very sharp distribution around a mean value for short- ranged correlations. The passage can be experimentally checked by tuning the external magnetic field for a ferromagnet at temperatures well below the Curie temperature.

V.   CONCLUSIONS

The probability distribution for the work done in taking a system from one thermodynamic state to another has often been studied analytically [7,12,19], but surprisingly the fluctuations around the average of the quantity $\Omega = \exp(-W / k_B T)$ has not been looked at before the investigation in Ref. [13] . There was general consensus that the probability distribution would be sharply centred around some average value $\bar{W}$ which would be of $O(N)$, where N is the system size, and would have a exponential ($\exp(-W / k_B T)$) tail for $W >> \bar{W}$ indicative of dominance of extreme events. The fact that a quantity like Ω, which is vanishingly small where the probability distribution peaks, can have very large fluctuations was generally overlooked. In Ref. [13], it was pointed out that this large fluctuation, as would follow from the explicit distributions in Refs. [12] and [19], is primarily a feature of the short-ranged correlations of the fluctuations. It was also pointed out that if the correlations were long ranged , as happens in NESS for driven fluids , then results would be substantially different. What was clarified in Ref. [28] was the fact that long- ranged correlations , whether coming from NESS in driven fluids or from Goldstone modes in ferromagnets at low temperatures, have the ability to set the scale of the work probability distribution to much lower values and thus provide a much broader distribution. For

$W \rightarrow 0$, the distribution $\rho(W) \rightarrow 0$ as $\rho(W) \propto \exp[-V(\ln\dfrac{\langle W \rangle}{W})^n / L^3]$, where n is a number

which is system specific. The scale is reduced from V to a potentially much smaller quantity V / L^3 where L can be several lattice spacings and thus for small systems at least, the fluctuations in Ω are significantly reduced, and when $L \square L_\perp$ it is very strongly reduced even for macroscopic systems. It would be interesting to revisit earlier experiments [36-41]

which confirmed or utilized the Jarzynski equality, Eq. (1.1), with the large fluctuations in mind .

Ferromagnets have the interesting feature that by tuning the external magnetic field from infinitesimal to finite values the correlations can be changed from long- ranged ones to short- ranged ones. Consequently, it would be the ideal system for studying the crossover in the fluctuations around the mean value that appears on one side of the Jarzynski equality [4]. We have provided the explicit forms of the probability distributions at small fields (long-ranged correlations) which means fields less than 100 Gauss in practical terms in Eqs. (4.18a) to (4.18c) and at fields which are larger than 100 Gauss (short ranged correlations) in Eqs. (4.28a) to (4.28c). The fluctuation is seen to be $O(\exp(\frac{L_\perp^2}{L^2}L_h^4))$ for small fields (small L_h^2). For larger magnetic fields, when the correlations are short ranged , this small W behaviour is severely suppressed and the fluctuation ε_Ω is enormous and $O(\exp(h^{3/2}L_\perp^2 L))$ as happens in other short ranged systems. The ferromagnet at low temperatures provide a unique opportunity to study the crossover shown in Eq. (4.21) by tuning the external magnetic field.

APPENDIX: AVERAGE WORK AND SHAPE OF THE PROBABILITY DISTRIBUTION

We want to discuss the consequences of the fact that the average, $<W>$, of the work done by the magnetization fluctuations is of $O(V)$, as seen from Eq. (3.4), even when the correlations are long-ranged making all the other moments of the work fluctuations proportional to $L_\perp^2 / L^2 = V / L^3$. In Sec IV, we saw the effect of this showing up in the construction of the work probability distribution $\rho(W)$ from the moment generating function $K(t)$. If we use only the $n=1$ and $n=2$ terms of the infinite series in Eq. (4.1), we get the Gaussian probability distribution shown in Eq. (4. 16a). If we want to calculate the tail of $\rho(W)$,then we need a closed form for $K(t)$ which can be obtained from the large-n terms of Eq. (4.1). The average value of W that one infers from this exercise is necessarily of $O(V / L)$. This Appendix describes how one interpolates the probability distribution from the very small values of W to values of the $O(V / L)$ and to even larger ones.

22

In the case of short- ranged correlations, as shown in Eq. (1.3), the average W is of $O(N)$ which is the same as $O(V)$ and the distribution is centred round this average. The distribution is also very sharp and a departure of $O(1)$ from the mean results in a suppression of the probability by the factor $O(\exp(-N))$. This is what causes the large value of ε_Ω, the fluctuation in $\exp(-W)$ which is almost negligible at the peak of the distribution .

For the long- ranged correlations we see that the distribution for $W \to 0$ is given by the structure (see Eq. (4.18a)), $\rho(W) \propto \exp\left(-\dfrac{L_\perp^2}{L^2}\left[\ln\dfrac{\bar{W}}{W}\right]^2\right)$, where the scale $\bar{W}$ is found to

be proportional to $L_\perp^2$ $(O(V/L))$ and the overall scale of the distribution is $L_\perp^2 / L^2 \propto V / L^3$. This form holds for $W << \bar{W}$ and produces a long-lived tail compared to the short-ranged case where the prefactor of the logarithm is proportional to V. We note that if $\bar{W}$ and W are of the same order, then the magnitude $\rho(W)$ is $O(\exp- (\dfrac{L_\perp^2}{L^2}))$. This is the same kind of probability distribution that one obtains from the $\rho(W)$ of Eq. (4.18b) . Thus the low- W form of Eq. (4.18a) merges into the Gaussian of Eq. (4.18b). However , using the Gaussian of Eq. (4. 16a) which gives the average of W correctly, we get a similar order of $\rho(W)$ only when $W - \langle W \rangle \approx 1 / L$.

Thus , the picture that emerges is that one has a Gaussian distribution which actually takes note of the fact that $\langle W \rangle$ is $O(V)$ and we can write this distribution as $\rho(W) \propto \exp(-L_\perp^2 (\dfrac{W}{<W>} - 1)^2)$ in the vicinity of $\langle W \rangle$. Clearly , if we consider values of $\dfrac{W}{\langle W \rangle} - 1$ which are $O(1/L)$, then the probability coming from such a distribution merges with the values obtained from the distribution found in Eq. (4.16) which in turn merges with the small W tail (as also the large W tail) in the appropriate limit. Given this, if we write the work probability distribution as

$$\rho(W) \propto e^{-L_\perp^2\left(\frac{W}{\langle W\rangle}-1\right)^2} \qquad \text{(A1)}$$

then

$$\left\langle W_{fl}^2\right\rangle = \left\langle \left(W - \langle W\rangle\right)^2\right\rangle = \langle W\rangle^2 / 2L_\perp^2 \qquad \text{(A2)}$$

Setting $\langle W \rangle = \sigma L_\perp^2 L$, where σ is a number of O(1), $\left\langle W_{fl}^2\right\rangle = \sigma^2 L_\perp^2 L^2 / 2$ and comparing with the exact answer of Eq. (3.14) we find that

23

$$\sigma^2 = \left(\frac{h}{MJ}\right)^2 \frac{1}{3\pi} \qquad\qquad \text{(A3)}$$

We can now evaluate the moments $\langle e^{-nW}\rangle$ and find

$$\mathcal{E}_\Omega = e^{\frac{1}{6\pi}\frac{L_\perp^2}{L^2}\left(\frac{hL^2}{MJ}\right)^2} = e^{\frac{1}{6\pi}AL_h^4} \qquad\qquad \text{(A4)}$$

in exact accordance with what was reported in Ref [28] and obtained in Eq. (4.21). This is also of the same form as in Eq. (4.24) , the only difference being in the prefactor of AL_h^4.

As for $W \gg \langle W\rangle$, we note that the tail expressed by Eq. (4.18c) gives a $\rho(W)$ of $O(\exp{-\left(\frac{L_\perp^2}{L}\right)})$. The distribution $\rho(W)$ of Eq. (A1) gives a result of same order for $\frac{W}{\langle W\rangle}-1 \approx O\left(\frac{1}{\sqrt{L}}\right)$. The above discussion supports the following picture for the work distribution function:

i) It is centred at W of $O(\langle W\rangle$ with $\langle W\rangle$ proportional to the volume of the system as shown in Eq. (A1) and its width is proportional to $L_\perp^2 L^2$.

ii) The tail of the distribution ($W \to 0$ *i.e.* $W \ll L_\perp^2$) is proportional to
$$\exp\left(-A\left(\ln\frac{1}{W}\right)^2\right)$$ and the distribution of Eq. (A1) merges into this tail for
$$\langle W\rangle - W > \frac{1}{L}$$

iii) For $W - \langle W\rangle > O(\frac{1}{\sqrt{L}})$, the distribution of Eq. (A1) merges with the large W tail of Eq. (4.18c)

The weakening of the mean square fluctuation is due to the presence of the prolonged $W \approx 0$ tail in the distribution function.

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