# Resonant Nonequilibrium Temperatures<sup>†</sup>

#### F. Ritort\*

Department of Physics, Faculty of Physics, University of Barcelona, Diagonal 647, 08028 Barcelona, Spain Received: October 10, 2004; In Final Form: November 28, 2004

In this paper, we investigate nonequilibrium temperatures in a two-state system driven to a nonequilibrium steady state by the action of an oscillatory field. The nonequilibrium temperature is determined by coupling a small cavity or probe to the nonequilibrium system and studying the fluctuating noise in the cavity, as has been proposed in the context of glassy systems. We show the presence of resonant effects in the nonequilibrium temperature and discuss the existence of a constitutive steady-state equation in such nonequilibrium conditions. We propose this simple model as an excellent system to carry out experimental measurements of nonequilibrium temperatures. This may help to better understand the physical meaning of such an elusive concept.

# 1. Nonequilibrium Temperatures

Nonequilibrium physics is a vast field of research plagued by many interesting open questions. A question that has attracted the attention of theorists for a long time is the possibility to define a nonequilibrium temperature<sup>1</sup> (NET). There is an important motivation behind the quest for such concept. One of the most important conceptual ingredients in classical thermodynamics is the notion of temperature. Temperature is a physical measure of the energy content of a system. When two bodies at different temperatures are put in contact with each other, heat flows from the body at higher temperature to the body at a lower temperature. This experimental fact allows the classification of thermodynamic states according to the value of their temperature (defined after establishing a suitable temperature scale). Bodies at identical temperatures do not exchange a net energy current when put in thermal contact.

The main goal when extending the concept of temperature to nonequilibrium systems is to establish under which conditions the previous experimental facts are still valid. But what apparently seems to be an easy promenade along the foothills of a new land turns out to be a fatal excursion full of traps and difficulties. It is not our purpose to enumerate here the casuistry of examples of nonequilibrium systems (many of them pretty simple and harmless) where such an extension has proved unsuccessful. Most of the difficulty lies in the transitivity property of temperature as well as in the fact that nonequilibrium bodies can be put in contact in many different ways, which do not necessarily lead to the same result. Yet, there is an important exception to all this complicated phenomenology, put forward by Onsager a long time ago. We refer to irreversible or nonequilibrium thermodynamics<sup>2</sup> in the regime where deviations from equilibrium are not large enough for currents (of heat, mass, energy, or charge) to be proportional to the externally applied forces. Sometimes this is also referred to as the linear response regime where fluctuations are Gaussian distributed. Linear systems are pretty interesting as they teach us that the NET must be a local concept.<sup>3</sup> For instance, in a wire of metal with the extremes in contact with two thermal sources, heat flows form the higher temperature source to the lower temperature one. The heat current is then proportional to the temperature gradient, and a temperature field (e.g., varying linearly along the wire) describes the nonequilibrium state of the system.

However, there is a more recent category of systems that deserve special attention because they seem to be good examples to investigate the concept of NET. These are aging or glassy systems that are prepared in an initial far from equilibrium state and slowly evolve toward equilibrium for astronomically large time scales. Under such conditions, a statistical interpretation of fluctuations can be quantitatively described by an extended fluctuation—dissipation theorem (FDT) introduced in the context of spin glasses. 4,5 As these systems slowly evolve toward equilibrium, they appear to thermalize over some regions of phase space and develop a restricted flat measure (we may refer to it as a *protomeasure*) similar to the microcanonical measure for equilibrium systems. The flat state reached by the system under such conditions defines a statistical effective temperature similar to that provided by statistical mechanics.<sup>6,7</sup> One important aspect of this NET is its dependence on the probed frequency as well as the age of the system. It has been suggested by Cugliandolo, Kurchan, and Peliti that such a pattern of NETs could be experimentally measured by using a small interacting probe that plays the role of a thermometer whose frequency can be tuned.<sup>8</sup> In the glassy jargon, it is common to denote this NET as effective temperature.

## 2. Motivation

Few experimental measurements of the NET exist. To date, the interpretation of most of these results still remains unclear. The first indirect experimental measure of NETs in aging systems dates back to magnetic noise measurements in spin glasses. In these measurements, systematic deviations for the fluctuation—dissipation theorem (FDT) were not observed, yet these did not probe the relevant frequency range. A few years ago, Israeloff and Grigera did voltage noise measurements in an electric resonant circuit formed by a capacitor containing glycerol and an inductance. Such experiments showed a faint dependence of the effective temperature on the age of the system. Subsequent Nyquist noise measurements by Ciliberto and co-workers on Laponite and polycarbonate have shown that the effective temperature is strongly dependent on frequency. At low enough frequencies ( $\omega t_w \ll 1$ , where  $t_w$  denotes the age

<sup>\*</sup> Author to whom correspondence should be addressed. E-mail: ritort@ffn.ub.es.

<sup>†</sup> Part of the special issue "David Chandler Festschrift".

of the system, also called waiting time), the effective temperature  $T_{\rm eff}(\omega,t_{\rm w})$  is up to 3 or 4 orders of magnitude larger than the bath temperature. Magnetization response and correlation measurements by the Ocio group in Paris vindicated strong FDT violations in spin glasses, and indeed these provided the first experimental determination of a fluctuation-dissipation (FD) plot. 12 More recently, the measure of NETs has shifted toward simpler systems and probes such as the stochastic motion of a torsional pendulum immersed in a granular media<sup>13</sup> or a bead joggling in a turbulent wind flow. <sup>14</sup> More intuitive results have been obtained in such cases. Optical tweezer measurements also recently have been used to study the fluctuating motion of a micrometer-sized bead immersed in an aging Laponite suspension. 15 No measurement of the effective temperature has to date provided a crystal-clear result of what theory predicts. There are fundamental questions about the effective temperature about which we do not yet know the answer. For instance, how is the measured temperature expected to depend on the characteristics of the probe (mass, frictional viscosity, or size)? Will the effective temperature depend on the type of observable measured?<sup>16</sup> Is the effective temperature a local or a global quantity? Ultimately, are the results reproducible when changing the experimental technique?

The situation is confusing and has some resemblance to other hot debates currently fashionable in the area of glasses. For example, regarding the question of heterogeneities, <sup>17</sup> are heterogeneities well-defined objects? Is there just one heterogeneity length scale or are there many? Does the measurement depend on the experimental technique?

The large amount of unanswered questions and the complexity of the problem demand going to simpler systems where specific predictions could be experimentally tested in a systematic way. In this paper, we propose the study of NETs in two-state-driven systems as a useful playground to elucidate many of the subtleties behind the concept of the NET. Several reasons compel us to consider such an illustrative example. On one hand, two-state systems represent the simplest examples of nonlinear systems with activated behaviors. On the other hand, they are physically realizable in many different examples in the laboratory and are easy to control. Finally, steady-state systems provide an interesting nonequilibrium regime characterized by non-Gibbsian distributions where several theoretical predictions can be tested. Our main goal here is to show that in such conditions NETs display resonant effects that are amenable to experimentation. The present study is by no means exhaustive; we just want to emphasize the importance of investigating simpler nonequilibrium systems and stimulate further experiments in this exciting area of research. The plan of the paper is as follows. In section 3, we describe the model for the nonequilibrium measurements. It consists of a nonequilibrium system plus an external probe or cavity used for the measurements. In section 4, we show the main results: existence of resonant effects in the NET and the possibility of defining a constitutive equation for the steady state. Finally, we discuss candidate systems where these effects could be experimentally tested.

### 3. Model

An interesting problem in statistical physics is to know whether it is possible to extend some of the concepts of thermodynamics to nonequilibrium systems in steady states (SS) and possibly characterize them by nonequilibrium constitutive equations. Nyquist noise measurements offer many potentialities as these are an ideal tool to probe thermal fluctuations around the steady state. Heat fluctuations are experimentally

accessible by using a resonant cavity coupled to the system. Although such measurements have been attempted in different contexts, experimental verifications are scarce, probably because of the great complexity of the systems addressed, <sup>10,11</sup> and experimental measurements turn out to be difficult. In this regard, numerical simulations become an alternative tool of research. Recently, Hatano and Jou<sup>19</sup> have investigated NETs in mechanically driven linear oscillators by coupling them to a resonant cavity. Here, we propose measurements of NETs in two-state systems (e.g., magnetic-nanoparticle or electric-dipole systems) coupled to a resonant probe and show the existence of resonant effects that are modulated by the physical properties of the cavity or probe. At the end, we suggest possible experiments that could check such predictions.

3.1. The System. A broad category of systems can be modeled by two-level systems. These offer realistic descriptions of electronic and optical devices that can function in two different configurations, atoms in their ground and excited states, magnetic particles whose magnetic moment can point in two directions, or biomolecules in their native and unfolded states, among others. In what follows and for sake of clarity, we will adopt the nomenclature of magnetic systems. A two-state unit has the magnetic moment  $\mu$  and can point in two directions according to the sign of the spin  $\sigma = \pm 1$ . In the presence of an external field H, the energy of the spin is given by  $E(\sigma) =$  $-\mu H\sigma$ . The transition rate for the spin will be denoted as  $p^{\text{up}}(H)$  and  $p^{\text{down}}(H)$  to indicate the transitions  $\sigma = -1 \rightarrow \sigma' =$ 1 and  $\sigma = 1 \rightarrow \sigma' = -1$ , respectively. These rates satisfy detailed balance; therefore,  $p^{up}(H)/p^{down}(H) = \exp(-2\beta\mu H)$ where  $\beta = 1/k_BT$ , with T being the bath temperature and  $k_B$ being the Boltzmann constant. The overall transition rate is given by  $p^{\text{tot}}(H) = p^{\text{up}}(H) + p^{\text{down}}(H)$ . We have chosen Glauber transition rates given by

$$p^{\text{up}}(H) = p^{\text{tot}}(H)q(H) \quad p^{\text{down}}(H) = p^{\text{tot}}(H)(1 - q(H)) \quad (1)$$

with  $q(H) = (1 + \tanh(\beta \mu H))/2$  and  $p^{\text{tot}}(H) = 1/\tau_{\text{relax}}(H) = \alpha(H)$  corresponding to the inverse of the relaxation time.

For the nonequilibrium state, we just consider that the spin is driven by an oscillating field of frequency  $\omega$  of the form  $H(t) = H_0 \cos(\omega t)$ . The transition rates for the spin are now time-dependent and given by

$$p^{\rm up}(t) = \alpha(H(t)) \frac{\exp(\beta \mu H(t))}{2 \cosh(\beta \mu H(t))}$$
 (2)

$$p^{\text{down}}(t) = \alpha(H(t)) \frac{\exp(-\beta \mu H(t))}{2 \cosh(\beta \mu H(t))}$$
(3)

For the relaxation time  $\tau_{\rm relax}(H)$  or  $\alpha(H)$ , we have chosen the barrier dependence mostly found in thermally activated magnetic systems (such as magnetic nanoparticles).<sup>20</sup> These are described by the Brown–Neel formula

$$\tau_{\rm relax}(H) = \tau_0 \exp\left(\frac{B(H)}{k_{\rm B}T}\right) \tag{4}$$

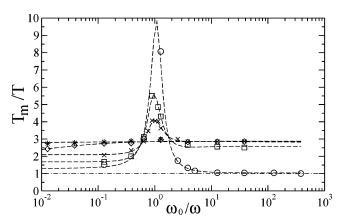
where  $\tau_0$  is a microscopic time describing relaxation within a state and B(H) is a field-dependent activation barrier. Related expressions exist for the case of a molecular bond that breaks under the action of a mechanical force.<sup>21</sup> In that case, the spin corresponds to the extension of the molecule, and the magnetic field is the applied force at the ends of the molecule. In the present study, we have considered the simplest case where  $B(H) = B_0$  is field-independent. This is a reasonable approxima-

tion. In superparamagnetic nanoparticles, the energy barrier is nearly field-independent (in contrast to ferromagnetic nanoparticles with uniaxial anisotropy where B(H) is not constant but depends on the intensity of the external field projected on the easy magnetization axis). Superparamagnetic nanoparticles can be experimentally realized in paramagnetic molecular clusters without a coercitivity field (yet, for these systems the magnetic signal of individual nanoparticles is expected to be very small). Other examples are specific ferro- and ferrimagnetic nanoparticles where the anisotropy contribution to the zero-field barrier is negligible (for a discussion see ref 22). The height of the barrier usually depends on the properties of the nanoparticle. Recent experiments have demonstrated how the height of the barrier  $B_0$  can be reduced (therefore  $\tau_{\text{relax}}(H)$  can be modulated), making it possible to observe magnetization-reversible transitions (also called telegraph noise measurements) in single cobalt nanoparticles at low temperatures.<sup>23,24</sup>

**3.2.** The Cavity. We model the resonant cavity as a harmonic oscillator with natural frequency  $\omega_0$ , mass m, and friction coefficient y (for an electrical cavity these correspond to a capacitance, an inductance, and a resistance, respectively) coupled to a bath at temperature T'. These parameters define an inertial time scale  $\tau_1 = 2\pi/\omega_0$  and a dissipative time scale  $\tau_{\rm dis} = \gamma/k$  where  $k = m\omega_0^2$  is the stiffness constant of the cavity. To model the interaction between the spin and the cavity, we add a term in the energy of the type  $-\epsilon \sigma x$  in  $E(\sigma) = -\mu H \sigma$ , giving  $E(\sigma) = -\mu H\sigma - \epsilon \sigma x$ , as well as in the equation of motion of the oscillator, where x describes the observable associated to the cavity (e.g., the voltage or charge of the capacitor). (This coupling can be interpreted in many ways. For instance,  $\sigma$  could stand for the magnetic  $(\vec{m})$  or electric  $(\vec{p})$  dipole moment of the system and x for the magnetic field (generated by an inductance) or the electric field (generated by a capacitor) of an electrical cavity. Therefore, the coupling would be linear of the form  $\vec{m}B$ or  $\vec{p}E$ , respectively. Moreover, to a first approximation, the magnetic field is proportional to the current intensity I, and the electric field is proportional to the voltage across the plates of the capacitor V. Therefore, x stands for either the current intensity I or voltage V (or charge Q) of the capacitor.) The cavity is governed by the stochastic equation

$$m\ddot{x} + \gamma \dot{x} + m\omega_0^2 x - \epsilon \sigma = \xi \tag{5}$$

where  $\xi$  is a white noise with correlations  $\langle \xi(t)\xi(s)\rangle =$  $2k_{\rm B}T'\gamma\delta(t-s)$ . Let the magnetic system be driven to a SS under the action of an oscillating external field  $H(t) = H_0 \cos(\omega t)$ , and let us put it in contact with the cavity as described previously. The average power supplied by the system upon the cavity is given by  $\mathcal{L} = \epsilon \sigma \dot{x}$  where the bar over the characters stands for the average over many cycles. If  $T' \gg T$ , then an average heat current will flow from the cavity to the system, and if  $T' \ll T$ , then the contrary will hold. For small enough  $\epsilon$ , the average power transferred between the probe and the system will be proportional to  $\epsilon^2$ . This result can be simply understood from symmetry considerations. By a change of  $\epsilon \rightarrow -\epsilon$  in eq 5, the dynamics of the probe are invariant under the spin inversion  $\sigma \rightarrow -\sigma$ . This last transformation also does not change the dynamics of the spin in the ac field. Hence, the transferred power  $\mathscr{D}$  must remain invariant under the transformation  $\epsilon \rightarrow$  $-\epsilon$ . Upon variation of T', there must be a temperature  $T_{\rm m} =$  $T'_{\omega,\omega_0,m,\gamma}(T)$  (which depends on the parameters of the cavity) at which the average power supplied to the cavity vanishes in a quadratic order in  $\epsilon$  (i.e.,  $\sigma \dot{x} = 0$  in a linear order in  $\epsilon$ ). Sometimes the net heat power exchanged between the system and the thermometer is considered,  $\mathcal{L} = \epsilon(\sigma \dot{x} - x\sigma)$  (where we



**Figure 1.** Measured values of the NET,  $T_m/T$ , for paramagnetic nanoparticles driven in an oscillating field. The parameters of the model are  $\mu = 300\mu_B$ , T = 200 K,  $H_0 = 2$  T,  $\tau_0 = 10^{-7}$  s, and  $B_0 = 2300$  K. The NET is determined by coupling the system to a cavity and finding the temperature of the cavity at which the average heat current between system and cavity vanishes. The cavity is characterized by different values of the resonant frequency  $\omega_0$  in the range 0.01-100 rad/s and a damping time scale  $\tau_{\rm dis} = \gamma/k = 1$  s. Different curves correspond to oscillating magnetic fields of intensity  $H_0 = 2$  T and different frequencies  $\omega \tau_{\text{relax}} = 0.001$  (circles), 0.01 (squares), 0.02 (crosses), 0.1 (diamonds), and 1(asterisks). The lines are the spectral line shape fits (see text).

take  $\dot{\sigma}(t) = \sigma(t+1) - \sigma(t)$ ). Note that from the relation  $\dot{x}\sigma =$  $\dot{x}\sigma + x\dot{\sigma}$  there is no difference between this and our definition as they coincide (except by a global sign) for SS systems. Only for non-SS systems (e.g., aging systems), would we expect to see differences between both quantities.<sup>25</sup>

### 4. Results

We have numerically integrated the stochastic equations (eqs. 3 and 5) under different conditions. The dynamics of the spin (eq 3) have been simulated by taking into account its interaction with the cavity. Note that when solving the coupled dynamics the system and the cavity feel the  $\mathcal{O}(\epsilon)$  interaction with their corresponding partner. Both the interaction of the system with the cavity and vice versa contribute to the total transferred power  $\mathcal{L}$  in the second order of  $\epsilon$ .

The number of parameters that we can vary in the simulations is very large. At first, there seem to be four different time scales  $\tau_{\rm relax}$ ,  $1/\omega$ ,  $\tau_{\rm dis}$ , and  $1/\omega_0$ . In what follows, we show results for a fixed value of the dissipative time scale of the cavity  $\tau_{\rm dis}$  =  $\gamma/k$ . However, out of the three remaining time scales, we are only left with just two independent variables. The argument is as follows. For a given value of the frequency of the ac driving field  $\omega$  and a value of the relaxation time of the spin  $\tau_{\rm relax}$  (eq 5), the results only depend on the adimensional product  $\omega \tau_{\rm relax}$ as one time of the two time scales  $(1/\omega \text{ or } \tau_{\text{relax}})$  can be used to rescale the time in the equation of motion for the spin in the SS. For a fixed value of  $\omega \tau_{\rm relax}$ , the value of  $T_{\rm m}$  depends only on the ratio  $\omega_0/\omega$  between the frequency of the oscillator cavity  $\omega_0$  and the ac driving field  $\omega$ . Again, another time scale ( $\omega_0$  or  $\omega$ ) can be rescaled in the equation of motion for the oscillator. Therefore, after having fixed the value of  $\tau_{\rm dis}$ , we are just left with the two adimensional quantities  $\omega au_{\rm relax}$  and  $\omega_0/\omega$  as free parameters. For each pair of the values  $\omega \tau_{\rm relax}$  and  $\omega_0/\omega$ , we have determined the value of  $T_{\rm m}$  for which the average power supplied to the cavity vanishes. The results obtained are shown in Figure 1 as the ratio  $\omega_0/\omega$  changes by several orders of magnitude (each curve corresponding to a value of  $\omega \tau_{\rm relax}$ ). The following parameters were chosen for the two-state system:  $\tau_{\rm dis} = 1 \text{ s}, \ \tau_0 = 10^{-7} \text{ s}, \ B_0 = 2300 \text{ K}, \ {\rm and} \ T = 200 \text{ K},$ 

TABLE 1: Parameters for the Fit to the Spectral Line Shape  $(Eq 6)^a$ 

$\omega  au_{ m relax}$	0.001	0.01	0.02	0.1	1
α	1.74	2.48	2.81	0.15	0.14
$\beta$	0	3.33	3.24	0.25	
a	1.79	1.68	2.10	0.98	2.87
b	1.04	2.59	2.88	5.45	-0.02
c	0.30	0.76	1.21	2.25	

<sup>a</sup> For the largest value of  $ωτ_{\rm relax} = 1$ , data have been fitted to the function  $T_{\rm m}(\bar{\omega})/T = a + b\bar{\omega}^{-2\alpha}$ . For  $ωτ_{\rm relax} = 0.1$ , the results of the fit are only approximate (see the discussion in section 4).

corresponding to a relaxation time of the spin  $\tau_{\rm relax} = 0.01$  s as given in eq 4.

We note several salient features in this figure:

- 1. Resonance effect. There is a resonant peak in  $T_{\rm m}/T$  for  $\omega=\omega_0$  that gets more sharp as the equilibrium regime  $\omega\tau_{\rm relax}\to 0$  is approached. On the contrary, if  $\omega\tau_{\rm relax}$  increases, then the resonance effect fades away. This effect can be easily understood. When  $\omega\tau_{\rm relax}\to 0$ , the system is driven slowly enough for the driven process to be quasi static or reversible. In this regime, the amount of dissipated heat (defined as the average dissipated work<sup>26</sup> along a cycle of the field) decreases proportionally to the ratio  $\omega\tau_{\rm relax}$ , and mechanical resonance is observed.
- 2. Spectral line shape. The value of  $T_{\rm m}/T$  becomes independent of the frequency of the cavity for large enough values of  $\omega_0$ .  $T_{\rm m}$  values can be well fitted to a spectral line shape of the form

$$T_{\rm m}(\bar{\omega})/T = (a + b\bar{\omega}^{2\alpha})/((\bar{\omega}^{\alpha} - 1)^2 + c\bar{\omega}^{\beta}) \tag{6}$$

with  $\bar{\omega}=\omega_0/\omega$  and  $\alpha$  and  $\beta$  exponent values satisfying  $\beta<2\alpha$ . The fit works well for not too large values of  $\omega\tau_{\rm relax}$  when the resonance peak is observed. For large values of  $\omega\tau_{\rm relax}$  (Figure 1), the curve  $T_{\rm m}(\bar{\omega})/T$  becomes flat (no resonance peak is observed), and the fit is only an approximated guess. In the limit  $\omega\to0$ , the right side of the spectral line shape converges to a Lorentzian plus a constant (equal to 1). The horizontal dot—dashed line  $T_{\rm m}/T=1$  corresponds to the equilibrium measurement that is only achieved for  $\bar{\omega}\gg1$  and  $\omega\to0$ .

3. Nonequilibrium temperature (NET)  $T_{\rm m}^*$ . A unique value of  $T_{\rm m}/T$  describes the SS at all frequencies (asterisks in the figure) corresponding to the strongly dissipative regime where  $\omega \tau_{\rm relax}$  is very large. Moreover, the quantity

$$T_{\rm m}^* = \lim_{\omega_0 \to \infty} T_{\rm m}(\omega_0) \tag{7}$$

has been shown (for instance in the case of weak turbulence<sup>27</sup> or in slowly relaxing systems<sup>8</sup>) to satisfy the Nyquist formula

$$T_{\rm m}^* = \lim_{\omega_0 \to \infty} \frac{\pi}{2} \frac{\omega_0 \tilde{C}_{\rm cavity}(\omega_0)}{\tilde{\chi}_{\rm cavity}(\omega_0)}$$
(8)

where  $\tilde{C}_{\text{cavity}}$  and  $\tilde{\chi}_{\text{cavity}}$  stand for the power spectrum and the out-of-phase susceptibility of the cavity, respectively. The value of  $T_{\text{m}}^*$  is therefore experimentally measurable by measuring the power spectrum of the cavity. (According to the fit formula (eq 6), the value of  $T_{\text{m}}^*/T$  should be equal to b in the limit  $\bar{\omega} \rightarrow \infty$ . However, by comparing the results of the fit shown in Table 1 with the results of Figure 2, we find qualitative agreement only for  $\omega \tau_{\text{relax}} \ll 0.1$  when the resonance peak is observed (for  $\omega \tau_{\text{relax}} = 0.1$ , the value of b is 5.45, too far from the expected value of approximately 2.8). This suggests that eq 6 is just an approximate fit to the data shown in Figure 1.)

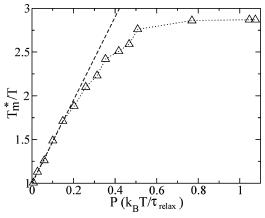


Figure 2. Constitutive equation of the magnetic system in its SS. P stands for the entropy production per particle of the magnetic system. The entropy production is expressed as the dissipated heat or work (the area enclosed in the M-H hysteresis cycle in  $k_BT$  units) divided by the relaxation time of the particles  $\tau_{\rm relax}$  as given in eq 4. The dashed straight line is the linear fit  $P = \alpha(T_{\rm m}^*/T - 1)$  with  $\alpha \simeq 0.2$ . The dotted line passing through the points is a guide to the eye. The same parameters are used as in Figure 1.

**4.1. Constitutive Steady-State Equation.** The NET  $T_m^*$  was determined in numerical simulations of a forced linear oscillator and shown to be dependent on the type of coupling between system and cavity.<sup>19</sup> For the two-state or spin system, we have verified this result by changing the type of coupling between oscillator and system. When the coupling energy term is changed from  $-\epsilon \sigma x$  to  $-\epsilon \sigma x^2$ , the value of  $T_{\rm m}$  changes noticeably (data not shown). Yet the value of  $T_{\rm m}^*$  can be a useful quantity to characterize the SS. As we said previously, the spin dissipates heat as it tries to follow the oscillations of the field. The amount of heat lost increases as the driving frequency  $\omega$  increases. The amount of heat is given by the area enclosed in the hysteresis cycle M - H of the system. A quantity that characterizes the nonequilibrium state is the entropy production P (i.e., the average dissipated heat per unit time). In extended irreversible thermodynamic theories,<sup>28</sup> the entropy production is expected to be a function of the previous NET,  $T_{\rm m}^*$ . This relation defines a constitutive steady-state equation. However, at difference with classical thermodynamics, this relation is expected to have some degree of ambiguity. Indeed, we have seen that the value of  $T_{\rm m}$ depends on the type of coupling; therefore a unique relationship (cavity independent) between the entropy production P and  $T_m^*$ cannot be established in this simple example for all types of couplings. In Figure 2, we show the SS constitutive equation found in our model; the value of  $T_{\rm m}^*$  grows linearly with the heat power and eventually saturates to a maximum value  $(T_{\rm m}^*/T \simeq 2.8(1))$  in Figure 2). The linear behavior found in the limit  $P \rightarrow 0$  (dashed line shown in Figure 2) can be expressed as  $P = \alpha (T_m^*/T - 1)$  where  $\alpha$  is a constant. We remark that this linear dependence is unconventional. In Onsager theory, the heat flux or current j between two bodies at different temperatures is governed by the Fourier law,  $j = k\nabla T$  where k is the thermal conductivity of the system. The entropy production P though is proportional to  $(\nabla T)^2$ . Accordingly, one might expect a quadratic (rather than linear) dependence for the dissipated power P with the difference  $(T_{\rm m}^* - T)$ , contrary to what is observed in Figure 2. As the dissipated power is always positive (as implied by the second law of thermodynamics), the linear dependence suggests that the measured temperature  $T_{\rm m}$  can never be lower than the bath temperature. This is in agreement with our findings; we could not find values for the parameters of the model such that  $T_{\rm m}$  was lower than T. The physical significance of the inequality  $T_{\rm m} > T$  in the context of nonequilibrium steady states remains unclear to us. (In this regard, it would be interesing to clarify whether this inequality is somehow related to an inequality describing transitions between nonequilibrium steady states put forward by Oono and Paniconi<sup>29</sup> in the context of the steady-sate thermodynamics. This inequality has been obtained as a particular case of a more general nonequilibrium equality derived by Hatano and Sasa<sup>30</sup> and experimentally tested very recently.<sup>31</sup>)

#### 5. Discussion

A few years ago, motivated by theoretical studies in spin glasses and structural glass models, Cugliandolo, Kurchan, and Peliti<sup>8</sup> proposed to define nonequilibrium temperature (NET) as the temperature that a cavity coupled to the system would measure. The measured temperature could be determined by putting the cavity in contact with its own thermal bath and modulating the temperature of that bath until the net heat power exchanged between the system and the cavity vanishes. In the limit of large cavity frequencies, such a temperature was found to coincide with the temperature obtained from the extended form of the FDT. Yet clear experimental evidence and simple experimental tests of this nonequilibrium temperature are yet to come.

In this paper, we have studied a simple nonequilibrium steadystate system that can be described by a nonequilibrium temperature (NET). It consists of a two-state system driven by an oscillating ac field and coupled to a cavity. Under general conditions, we predict the existence of a resonant peak for the NET at values of the driving frequency  $\omega$  that are equal to the resonant frequency of the cavity  $\omega_0$  (Figure 1). For large values of  $\omega_0$ , the measured temperature asymptotically converges to a well-defined value that is a function of the dissipated power, allowing us to define a constitutive equation for the steadystate system. The origin of this resonance effect is mechanical; the frequency of the cavity  $\omega_0$  resonates with the frequency of the ac field  $\omega$ . Although we did not study it in detail, a similar resonant effect is predicted in the limit of a purely overdamped cavity  $(\omega_0 \rightarrow 0)$  when the relaxational frequency of the cavity  $\omega_{\rm dis} = 1/\tau_{\rm dis} = k/\gamma \approx \omega$ . The origin of this resonance would then be stochastic. This phenomenon would then fall in the category of stochastic resonance phenomena,<sup>32</sup> an ubiquitous behavior found in relaxational systems.

The results of Figures 1 and 2 could be experimentally tested in magnetic or dielectric systems. For instance, in the case of dielectric systems, an oscillating voltage of intensity  $V_0$  and frequency  $\omega$  could be applied between the metallic plates of a capacitor containing a dielectric material with relaxational electric dipoles that have a characteristic time scale  $\tau_{\rm relax}$ . The dipolar charge in the surface of the capacitor is then coupled to an external RLC circuit (of resistance R, capacity C, and inductance L) that acts as a cavity. The cavity has characteristic resonant frequency  $\underline{\omega}_0 = 1/\sqrt{LC}$  and a damping frequency  $\omega_{\rm dis} = 1/\tau_{\rm dis} = 1/\sqrt{RC}$ . For a fixed frequency of the ac electric voltage  $\omega$  and the frequency of the cavity  $\omega_0$ , one could then determine the voltage noise density and the electric impedance of the cavity from which one could obtain values for the measured  $T_{\rm m}$  by using the generalized Nyquist relation (eq 8). 10,11 This also would allow one to measure the dissipated power of the cavity  $P = V^2/|Z|$  (Z being the electric impedance) as a function of  $T_{\rm m}$ . Reproducing the qualitative results of Figures 1 and 2 should be within reach with current experimental techniques.

The results of Figures 1 and 2 could be also experimentally tested in magnetic systems (e.g., nanoparticles or paramagnetic systems) by coupling the ac magnetization signal to a resonant electric cavity and measuring the voltage noise density at different frequencies. This technique has been used to measure magnetostochastic resonance<sup>32,33</sup> in ferrite—garnet films using the Faraday effect. Another tool could be provided by transverse-field ac measurements.<sup>34</sup>

Let us mention that results quite similar to those shown in Figure 1 have been recently reported in the measurement of effective temperatures in a colloidal suspension of aging Laponite by using the nonequilibrium Einstein relation. The control parameter there is the age of the system  $t_{\rm w}$  rather than the driving or probing frequencies ( $\omega$  and  $\omega_0$ , respectively). In these measurements, the maximum of the temperature was interpreted as a resonance between the probe frequency  $\omega_0$  and the inverse of the relaxation time scale of the suspension ( $\tau_{\rm relax}$  in the present case).

To conclude, given the elusive interpretation of the concept of effective temperature and the formidable complexity of glassy systems, it would be very instructive to carry out systematic research of NETs in simple noninteracting systems driven to nonequilibrium steady states. A simple experiment such as the one that we have described should not be difficult to carry out and would help us to better understand what we can expect from noise measurements in more complex nonequilibrium systems. In this respect, two-level systems are simple model systems where theoretical predictions for the NETs could be experimentally tested at both qualitative and quantitative levels.

**Acknowledgment.** I am grateful to David Jou for a critical reading of the manuscript. I also acknowledge support from the Spanish Ministerio de Ciencia y Tecnología Grant BFM2001-3525, Generalitat de Catalunya, the STIPCO network, and the SPHINX program.

### **References and Notes**

- (1) Casas-Vazquez, J.; Jou, D. Rep. Prog. Phys. 2003, 66, 1937.
- (2) De Groot, S. R.; Mazur, P. Non Equilibrium Thermodynamics; Dover Publications: New York, 1984.
  - (3) Vilar, J. M.; Rubí, M. Proc. Natl. Acad. Sci. U.S.A. 201, 98, 11081.
  - (4) Cugliandolo, L. F.; Kurchan, J. Phys. Rev. Lett. 2003, 71, 173.
- (5) For a recent review see: Crisanti, A.; Ritort, F. *J. Phys. A: Math. Gen.* **2003**, *36*, R181. Cugliando, L. F. *Les Houches*, Session 77; Barrat, J. L., et al., Eds.; Springer-Verlag: Berlin, 2003.
- (6) Ritort, F In *Unifying Concepts in Granular Media and Glasses*; Coniglio, A., et al., Eds.; Elsevier: Amsterdam, 2004; p 129 (preprint cond-mat/0311370).
- (7) The robustness of the concept of thermodynamic temperature has its roots in the statistical interpretation of heat provided by statistical mechanics. According to it, temperature is directly related to the gradient in the density of energy levels  $\rho(E)$ ,  $1/T = \partial S(E)/\partial E$  where  $S(E) = k_{\rm B} \log(\rho(E))$ . The statistical temperature is identical to the thermodynamic temperature because two bodies at mutual equilibrium reach a maximal entropy state compatible with the total energy available.
- (8) Cugliandolo, L. F.; Kurchan, J.; Peliti, L. *Phys. Rev. E* **1997**, *55*, 3898.
- (9) Alba, M.; Hammann, J.; Ocio, M.; Refregier, P.; Bouchiat, H. J. Appl. Phys. 1987, 61, 3683.
  - (10) Grigera, T. S.; Israeloff, N. E. Phys. Rev. Lett. 1999, 83, 5038.
- (11) Bellon, L.; Ciliberto, S.; Laroche, C. Europhys. Lett. 2001, 53, 511; Buisson, L.; Bellon, L.; Ciliberto, D. J. Phys.: Condens. Matter 2003, 15, \$1163
  - (12) Herisson, D.; Ocio, M. Phys. Rev. Lett. 2002, 88, 257202.
- (13) D'Anna, G.; Mayor, P.; Barrat, A.; Loreto, V.; Nori, F. *Nature* **2002**, *424*, 1909.
- (14) Ojha, R., P.; Lemieux, P. A.; Dixon, P. K.; Liu, A.; Durian, D. J. *Nature* **2004**, *427*, 521.
- (15) Abou, B.; Gallet, F. Probing a Non-Equilibrium Einstein Relation in an Aging Colloidal Glass. *Phys. Rev. Lett.* 2004, 93, 160603.
  - (16) Fielding, S. M.; Sollich, P. Phys. Rev. Lett. 2002, 88, 050603.
  - (17) Ediger, M. D. Annu. Rev. Phys. Chem. 2000, 51, 99.

- (18) Sasa, S.; Tasaki, H. Steady-State Thermodynamics for Heat Conduction. Unpublished work (preprint cond-mat/0108365).
  - (19) Hatano, T.; Jou, D. Phys. Rev. E 2003, 67, 026121.
  - (20) Wernsdorfer, W. Adv. Chem. Phys. 2001, 118, 99.
  - (21) Evans, E. Annu. Rev. Biomol. Struct. 2001, 30, 105.
  - (22) Gonzalez-Miranda, J. M.; Tejada, J. Phys. Rev. B 1994, 49, 3867.
- (23) Wernsdorfer, W. et al. J. Magn. Magn. Mater. 1995, 145, 33. Wernsdorfer, W.; et al. Phys. Rev. Lett. 1997, 78, 1791.
  - (24) Jamet, M.; et al. Phys. Rev. Lett. 2001, 86, 4676.
  - (25) Garriga, A.; Ritort, F. Eur. Phys. J. B 2001, 21, 115.
  - (26) Ritort, F. J. Stat. Mech: Theor. Exp. 2004, P10016

- (27) Hohenberg, P.; Shraiman, B. Physica D 1989, 37, 109.
- (28) Jou, D.; Casas-Vazquez, J.; Lebon, G. Extended Irreversible Thermodynamics, 3rd ed.; Springer: Berlin, 2003.
  - (29) Oono, Y.; Paniconi, M. Prog. Theor. Phys. Suppl. 1998, 130, 29.
- (30) Hatano, T.; Sasa, S. *Phys. Rev. Lett.* **2001**, *86*, 3463. (31) Trepagnier, E. H.; Jarzynski, C.; Ritort, F.; Crooks, G. E.; Bustamante, C. J.; Liphardt, J. Proc. Natl. Acad. Sci. U.S.A. 2004, 101,
  - (32) Gammaitoni, L.; et al. Rev. Mod. Phys. 1998, 70, 223.
  - (33) Grigorenko, A. N.; et al. J. Appl. Phys. 1994, 76, 6335.
  - (34) Spinu, L.; et al. J. Appl. Phys. 2000, 87, 5490.