RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 35, NUMBER 3 15 JANUARY 1987-II

Ubiquity of logarithmic scaling, $1/f$ power spectrum, and the $\pi/2$ rule

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(Received 22 October 1986)

For disordered systems governed by activated processes with a broad distribution of barrier heights, the susceptibility should obey logarithmic frequency scaling, and the associated noise power spectrum should be of the $1/f$-type (with logarithmic corrections). Further, various "$\pi/2$ rules" can be derived from the Kramers-Kronig relationships. The results are valid both near $T=0$ and near a phase transition when the latter exists. Two new examples of this very general behavior are discussed: the thermal properties of ordinary glasses and the impurity conduction and dielectric response of insulators.

INTRODUCTION

The response of many real systems to external perturbations should be governed by a broad distribution of relaxation times. This distribution may be due to a variation in the height of the barriers that the constituent units (ions, molecules or molecular orientations, electric dipoles, spins or spin clusters, electrons) have to cross in order to effect the response to the external field. It is natural to expect a rather broad distribution of such barriers in a disordered system where the barrier is determined by some local arrangement of the elements of the systems. Well-known examples are magnetic clusters in disordered magnets (e.g., spin glasses, random field systems), two-level systems and similar phenomena in ordinary glasses, defect motion in ordinary crystals, and, e.g., donor excitation energies in semiconductors or insulators. The latter may not be uniform, due to changes in the local environments of the donors and/or donor-donor interactions that depend on the particular configuration of neighboring donors, etc.

Such a distribution of relaxation times has been known to produce, in special cases, some interesting phenomena, such as logarithmic scaling of the response function in spin-glasses,[1] with resulting approximate $1/f$-type power spectrum of the associated fluctuations[1,2] and certain relationships between the real and imaginary parts of the response function.[3,4] Earlier, $1/f$-type fluctuations in conductors have also been shown to possibly be due to distributions in the activation barriers limiting the motion of some defects.[5] It is the purpose of this Rapid Communication to point out the *extreme generality* of these effects. They should exist in *almost any system with some randomness.*

After reviewing briefly the existing theoretical ideas, generalizing from the cases of $1/f$ noise and disordered magnets, we shall analyze along those lines two totally unrelated examples: the time-dependent specific heat of ordinary glasses and the frequency-dependent dielectric constant and conductivity associated with impurity conduction of insulators. To the best of our knowledge, these have never been analyzed in this way. We find that a surprisingly good description of the above two cases is provided by the above picture. The fit to many trends in the data is better than that of popular phenomenological forms which have previously been used to discuss these data, usually with no theoretical motivation.

We propose that this picture should be applicable to many relaxation and response phenomena (e.g., dielectric, thermal, magnetic, ultrasonic...) in disordered systems, and that one should try to both fit much of the existing data to these ideas and to obtain new data.

Finally, we shall discuss the low temperature versus $T\rightarrow T_c$ scaling and mention some possibly related situations such as variable-range hopping conduction at finite frequencies.

SHORT REVIEW OF THEORETICAL IDEAS

In the case of a simple relaxation model the noise spectrum of a system governed by a relaxation time τ is given by[5]

$$S(\omega)=\frac{1}{\pi}\frac{1/\tau}{\omega^2+(1/\tau)^2} . \tag{1}$$

Consider the case where τ is thermally activated

$$\tau(\Delta)=\tau_0 e^{\Delta/k_BT} . \tag{2}$$

Let us suppose that the system has a distribution of activation energies given by a (normalized) distribution function $P(\Delta)$. The total spectrum of the system will be given by

$$S(\omega)=\int_0^\infty P(\Delta)S(\Delta,\omega)d\Delta . \tag{3}$$

One is usually interested in a certain range of ω, say $\omega_1\lesssim\omega\lesssim\omega_2$, hopefully containing many decades, where, typically, $\omega\tau_o\ll 1$. The relevant range of Δ is given by

$$|\ln(\omega_2\tau_0)|\ll\frac{\Delta}{k_BT}\ll|\ln(\omega_1\tau_0)| . \tag{4}$$

The function

$$S(\Delta,\omega)=\frac{\tau_0\exp(\Delta/k_BT)}{1+\omega^2\tau_0^2\exp(2\Delta/k_BT)} \quad (5)$$

is a strongly peaked function of Δ with a width of order k_BT. If $P(\Delta)$ is a smooth function, it is reasonable to assume that it does not vary much in the range of $k_BT\ln(\omega_2/\omega_1)$ (usually $\Delta\sim 1$ eV, while k_BT is ~ 300 K or much less). Thus we can factor out $P_0=P(\Delta_0)$ where $\Delta_0=-k_BT\ln(\omega\tau_0)$ is the value of Δ at which $S(\Delta,\omega)$ peaks. The remaining integral is elementary, and we obtain[5]

$$S(\omega)\propto\frac{k_BT}{\omega}P(-k_BT\ln(\omega\tau_0)) \ . \quad (6)$$

From the fluctuation-dissipation theorem,

$$S(\omega)=\frac{k_BT}{\omega}\chi''(\omega) \ , \quad (7)$$

we obtain $\chi''(\omega)$, the imaginary part of the associated susceptibility. It follows that a broad distribution of activation barriers leads to $1/f$ noise (with logarithmic corrections) and to susceptibilities which exhibit logarithmic frequency dependences,

$$\chi''(\omega)=\chi''(-k_BT\ln(\omega\tau_0)) \ . \quad (8)$$

Such $1/f$ noise has been observed in the ordered phase of spin glasses.[1,2]

It should be emphasized that the particular form of $S(\omega)$, Eq. (1), is not important as long as $S(\omega,\Delta)$ is a strongly peaked function once we have substituted Eq. (2) for $\tau(\Delta)$.

$\pi/2$ RULES

The real and imaginary parts of the susceptibility are related by the Kramers-Kronig relations, e.g.,

$$\chi'(\omega)=\frac{1}{\pi}\int_{-\infty}^{\infty}d\omega'\frac{\chi''(\omega')}{\omega'-\omega} \ . \quad (9)$$

If $\chi''(\omega)$ is taken to have the form of Eq. (8) it is convenient to change variables with $y=-k_BT\ln(\omega'\tau_0)$ and $z=-k_BT\ln(\omega\tau_0)$. Making use of the fact that $\chi''(\omega)$ is an odd function of frequency, Eq. (9) then becomes[6]

$$\chi'(z)=\frac{1}{\pi}\frac{1}{k_BT}\int_{-\infty}^{\infty}dy\,\chi''(y)\left[\frac{1}{e^{(y-z)/k_BT}+1}-\frac{1}{e^{(y-z)/k_BT}-1}\right] \ . \quad (10)$$

In the limit $k_BT\rightarrow 0$ both terms in the integrand factor in brackets effectively approach step functions. Then, taking a derivative with respect to $z\propto\ln\omega$ one finds,

$$\chi''=-\frac{\pi}{2}\frac{d\chi'}{d\ln\omega} \ . \quad (11)$$

The condition for the validity of Eq. (11) is that χ'' vary slowly over the widths of the "step functions" in Eq. (10). This "$\pi/2$ rule" relating the real and imaginary parts of the susceptibility was first derived by Lundgren, Svedlindh, and Beckman[3] for the ordered phase of spin glasses. It was obtained (as in the derivation above) on the assumption that the probability distribution of relaxation times in a spin glass is slowly varying in $\ln\tau$. In Ref. 5 this $\pi/2$ rule was derived, not in the limit $T\rightarrow 0$ but in the limit $T\rightarrow T_c$, in the case of a phase transition in a disordered system with transition temperature T_c. An example is the random field Ising model for which the spin-relaxation time near T_c is believed to behave as[7,8]

$$\tau\sim e^{(c\xi^\theta)} \ , \quad (12)$$

where ξ is the correlation length and θ a dynamic critical exponent.[9] That is, in this case the effective barrier $W=c\xi^\theta\rightarrow\infty$ as $T\rightarrow T_c$, whereas for the derivation presented above the effective barrier, $W=\Delta/k_BT$ approaches ∞ as $T\rightarrow 0$. If in the phase-transition case we similarly assume a broad distribution of relaxation times, one obtains the logarithmic scaling form[7,8]

$$\chi''(\omega)=\chi''(-\ln\omega/\xi^\theta) \ , \quad (13)$$

completely analogous to Eq. (8) except that k_BT is replaced by $1/\xi^\theta$.

Thus, a logarithmic scaling form is obtained near T_c in phase transitions in disordered systems with c in Eq. (12) playing the same role as Δ in Eq. (2). The logarithmic frequency dependence may, however, be obtained much more generally at low temperature in disordered systems, whether or not such systems undergo phase transitions.

In addition to the $\pi/2$ rule given by Eq. (11), there is an analogous relationship involving the temperature derivative[3]

$$\chi''=-\frac{\pi}{2}\frac{1}{\ln(\omega\tau_0)}\frac{d}{dT}(\chi'T) \ . \quad (14)$$

The analogue of this low-temperature relationship near a phase transition with activated scaling (such as the random field Ising model) is

$$\chi''=\frac{\pi}{2}\frac{1}{\ln(\omega\tau_0)}(\xi^\theta)^2\frac{\partial}{\partial\xi^\theta}\left[\frac{\chi'}{\xi^\theta}\right] \ . \quad (15)$$

If we set $\xi=|T-T_c|^{-\nu}$, this can be rewritten,

$$\chi''=-\frac{\pi}{2}\frac{1}{\ln(\omega\tau_0)}\left[\chi'+\frac{(T-T_c)}{\nu\theta}\frac{d\chi'}{dT}\right] \ , \quad (16)$$

the form of which is closely analogous to the low-temperature expression

$$\chi''=-\frac{\pi}{2}\frac{1}{\ln(\omega\tau_0)}\left[\chi'+T\frac{d\chi'}{dT}\right] \ . \quad (17)$$

In terms of conductivities in activated systems at low temperatures, the $\pi/2$ rules take the forms

$$\frac{\sigma'}{\omega}=\frac{\pi}{2}\frac{d}{d\ln\omega}\left[\frac{\sigma''}{\omega}\right] \ , \quad (18)$$

$$\frac{\sigma'}{\omega}=\frac{\pi}{2}\frac{1}{\ln(\omega\tau_0)}\frac{d}{dT}\left[T\frac{\sigma''}{\omega}\right] \ . \quad (19)$$

The relevant response functions here are (σ'/ω) and

(σ''/ω) because it is the susceptibilities, rather than the conductivities themselves, which are expected to vary slowly (logarithmically) as functions of ω.

APPLICATION TO ORDINARY GLASSES

Recently, some very pretty experiments have been performed[10] which measure the real and imaginary parts of the product $c_p\kappa$ in several glasses. Here c_p is the specific heat at constant pressure and κ the thermal conductivity. Glasses are expected to be well described in terms of activation over barriers with a broad distribution of their heights. Thus the ideas presented in this paper should be expected to apply to this case.

The data were analyzed in terms of a Williams-Watts function. However, the logarithmic derivative of the real part closely follows the imaginary part, as expected according to Eq. (11). For both glycol and glycerol where $\mathrm{Re}c_p\kappa$ and $\mathrm{Im}c_p\kappa$ were measured for three different temperatures we find the $\pi/2$ rule, Eq. (11), to be satisfied in all cases to within about 15%, even though the data are very far from the $T=0$ limit. This rule has previously been tested for spin[3,11] and dipolar[4] glasses where it was also found to be satisfied, at least approximately.

If we apply Eq. (14) to the temperature dependence of $\mathrm{Re}c_p\kappa$ and $\mathrm{Im}c_p\kappa$ we find a value of τ_0 which is much too small ($\ln\tau_0\approx -100$). If instead we use Eq. (16), we obtain a reasonable fit to the data using

$$\Delta=\left(\frac{T-T_g}{v\theta}\right)$$

as a fitting parameter. For $v\theta=1$ the fit gives the value $T_g\approx 180$ K for glycerol.

An earlier attempt to fit Eq. (14) in spin glasses gave, similarly, a value of τ_0 which was unreasonably small[3] ($\ln\tau_0\approx -230$). However, since the data were taken in the neighborhood of the spin-glass transition temperature, a fit to Eq. (16) instead may possibly yield a more reasonable value of τ_0.

IMPURITY CONDUCTION IN INSULATORS

The frequency dependence impurity conduction is found to be well described in many cases by a power law,

$$\sigma'=A'\omega^s \ , \tag{20}$$

$$\sigma''=A''\omega^s \ , \tag{21}$$

where σ' and σ'' are the real and imaginary parts of the conductivity and A' and A'' are constants. The exponent s is near one, and both $s<1$ and $s>1$ are observed experimentally.[12] We suggest that the appropriate forms for σ' and σ'' may be

$$\sigma'=A'\omega f'(-\ln\omega\tau_0) \ , \tag{22}$$

$$\sigma''=A''\omega f''(-\ln\omega\tau_0) \ , \tag{23}$$

such that (σ'/ω) and (σ''/ω) will have the requisite slow variation in ω to make the $\pi/2$ rule, Eq. (18), apply. We have looked at the classic paper of Pollak and Geballe.[13] As shown in this paper, a good description of the data was obtained either with the power-law fit, Eq. (20), with $s\approx 0.8$ or the form of Eq. (22) with $f'=(-\ln\omega\tau_0)^4$ and $\tau_0=1.410^{-13}$. The latter form was also derived theoretically from a model with a distribution of relaxation times. We have tested the $\pi/2$ rule in the form of Eq. (18) and found this rule to hold quantitatively to within about 10% for the data presented in Ref. 13.

From the relationships

$$\frac{\varepsilon(\omega)}{\varepsilon_0}=1+4\pi\chi(\omega) \ , \tag{24}$$

$$\varepsilon(\omega)=1+\frac{4\pi}{\omega}i\sigma(\omega) \ , \tag{25}$$

it follows that

$$\frac{1}{\varepsilon_0}\frac{\sigma'(\omega)}{\omega}=\chi''(\omega) \ , \tag{26}$$

and the noise spectrum Eq. (7) takes the form

$$S=\frac{c_1k_BT}{\omega}(-\ln\omega\tau_0)^4 \ , \tag{27}$$

or for the power-law fit, Eq. (20),

$$S=c_2\frac{k_BT}{\omega^p} \ , \tag{28}$$

where $p=2-s$. We prefer the logarithmic form. We believe this is another example where activated processes give rise to logarithmic ω dependence and $1/f$-type noise. It is interesting to note that an exponent $s<1$ in the conductivity gives an exponent $p>1$ for the $1/f$ noise and vice versa. A related possible example is finite frequency hopping conduction which was argued[14] to produce a $\omega\ln\omega$ conductivity due to Coulomb interactions.

In conclusion we note that logarithmic scaling, $1/f$ noise, and various $\pi/2$ rules are all intimately related and should be observed in a large number of unrelated disordered systems both near phase transitions and, much more generally, at low temperatures, whether or not such systems undergo phase transitions. In addition to spin and dipolar glasses, and the random field Ising model, for which many of these ideas have been previously considered, we have briefly discussed here two additional examples: the thermal properties of ordinary glasses and the frequency-dependent conductivity of insulators. We suggest that the proper descriptions of these data are in terms of logarithmic frequency dependences (the Williams-Watts function has power-law frequency dependence for small ω, and the conductivity data are usually analyzed in terms of powers ω^s with $s\approx 1$). There should be associated $1/f$ noise behavior for appropriate correlation functions (with logarithmic corrections), and appropriate $\pi/2$ rules should be satisfied. We suggest that analyzing the data of many other disordered systems along these lines may be a fruitful approach.

ACKNOWLEDGMENTS

This research was partially supported by a grant from the Israeli Academy of Sciences. We are grateful to T. G. Castner for a discussion and to N. O. Birge for making a paper available prior to publication.

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