Evidence for the Importance of Interactions between Active Defects in Glasses

Sven Rogge, Douglas Natelson, and D. D. Osheroff
Department of Physics, Stanford University, Stanford, California 94305-4060
(Received 12 December 1995)

We report new evidence obtained between 500 μK and 150 mK for the importance of interactions between tunneling two level systems in structural glasses. After the application of a large dc electric field we observed logarithmic relaxations in $\epsilon'(\omega)$ and $\epsilon''(\omega)$ for up to 10^4 s. The frequency dependence of the relaxation in $\epsilon'(\omega)$ observed at higher temperatures vanishes for temperatures well below the minimum of $\epsilon'(\omega, T)$ and we find $\epsilon''(\omega) \propto T$ below the high temperature plateau, both qualitatively in agreement with models involving interactions.

PACS numbers: 61.43.Fs, 71.55.Jv, 77.22.Gm, 77.55.+f

For over 20 years our understanding of the anomalous universal properties of glasses at low temperatures, first recognized by Zeller and Pohl in 1971 [1], has been based on a model of independent tunneling two level systems (TLS), in which a broad distribution of thermally active defects in amorphous materials produces the heat capacity nearly linear in T and the thermal conductivity roughly proportional to T^2 seen in experiments. The measured exponents are closer to $T^{1.25}$ and $T^{1.9}$, however, and, as Lasjaunias, Maynard, and Vandorpe [2] have pointed out, attempts to correct these exponents by modifying the density of states of TLS cannot simultaneously account for both these differences. Recently, Yu and Leggett [3] have suggested that interactions between TLS mediated by their coupling to strain fields might resolve these and other discrepancies between theory and experiment.

In 1994 Salvino *et al.* [4] studied the ac dielectric response of structural glasses to large dc electric fields, and observed that a local minimum develops in $\epsilon'(\omega, E)$ after the application of any dc field. Models have been developed to explain the temperature and frequency dependence seen in these results in terms of the formation of a hole in the distribution of TLS at zero local field as equilibrium in the system is approached, owing to interactions between the TLS. It is these TLS which contribute most to the dielectric response, and hence the development of a hole in the TLS at zero field would produce the observed local minimum in $\epsilon'(\omega, E)$. Similar mechanisms are known to exist in spin glasses.

The existence of a hole in the density of TLS at zero local field might have significant effects on equilibrium properties of amorphous solids, and thus we have carried out a series of new experiments at much lower temperatures to further test this hypothesis. In this work, we find that the frequency dependence seen in $\epsilon'(\omega, E)$ disappears at low temperatures, in a manner expected from the models. In addition, we have observed that the temperature dependence of the equilibrium dissipation, $\epsilon''(\omega, T)$, at low temperatures varies linearly with temperature, contrary to what is expected for noninteracting TLS, but in accord with calculations based on a model including interactions. This suggests that the formation of a hole in the density of states can indeed affect equilibrium properties of amorphous systems.

One can attempt to describe the nonequilibrium dielectric response of TLS in amorphous solids to a dc field from two broad points of view: the interacting and noninteracting pictures. In the interacting picture Burin [5] considered TLS coupled to one another by a weak strain-mediated elastic dipolar interaction. He calculated the effect on an initially flat density of states as a function of local field due to this interaction and found that a hole in the density of states formed at zero local field. The depth of the hole grew logarithmically in time as the TLS approach thermal equilibrium. The appearance of such a feature is understandable when one considers the familiar stability argument used to explain analogous behavior in spin glasses [6]. One can explain the jump in the dielectric response due to the applied dc field in the following way: When the external dc field is changed, the density of TLS at zero local field is increased as thermal equilibrium is destroyed. The number of TLS at zero local field decreases as equilibrium is reestablished.

Putting this "dipole gap" into the density of states of the TLS, Burin used the equilibrium noninteracting TLS formalism [7] to calculate the dielectric response in the interacting picture. This procedure predicts a jump and subsequent relaxation in $\epsilon'(\omega)$ due to the application of the dc bias. The relaxation is frequency and temperature dependent at $T > T_{\min}$, but frequency independent for $T \ll T_{\min}$ due to the fact that only TLS relaxational tunneling, which disappears at low temperatures, is frequency dependent. Finally, the time dependent density of states also leads to a jump and logarithmic decay in the imaginary part of the dielectric response, $\epsilon''(\omega)$.

An approach similar to the above was employed by Carruzzo, Grannan, and Yu [8]. This group also performed a Monte Carlo simulation of a 3D nearest-neighbor Ising spin glass with 1000 spins to model interacting TLS. They found that the interactions cause a decreasing density of TLS at zero local field analogous to

 0031-9007/96/76(17)/3136(4)$10.00

the one discussed in [6]. This relaxation is logarithmic in time and is reflected as such in $\chi'(\omega)$. The temperature and frequency dependence of this relaxation is in good agreement with the $\epsilon'(\omega)$ relaxations presented in [4]. In contrast to an experiment performed on an RKKY spin glass [9] this model does not show a logarithmic relaxation in $\chi''(\omega)$.

Carruzzo, Grannan, and Yu also tried to explain the nonequilibrium dielectric response to the application of a large dc field by assuming noninteracting TLS. In this framework the equations of motion obeyed by the individual TLS are the Bloch equations (in the rotating wave approximation [10]). Thus the TLS are an analog to a spin system in a magnetic field (NMR) with a distribution of τ_1. This model predicts logarithmic relaxations in $\epsilon'(\omega)$ due to the rethermalization of the TLS after the bias application. The relaxations are temperature but not frequency dependent. Using the Landau-Zener criterion one can define two regimes showing different behavior, adiabatic and nonadiabatic, based on the switching time of the dc bias field compared to the tunneling and relaxation time of a given TLS. Since TLS have a broad distribution of tunneling times, for a given dc bias rise time some fraction of the TLS will always be in the nonadiabatic regime. In the adiabatic regime this model predicts a jump upward in $\epsilon'(\omega)$ and a subsequent logarithmic relaxation. A completely nonadiabatic response produces a jump downward in $\epsilon'(\omega)$ and an upward relaxation back to equilibrium. The experimental observation should be a combination of both responses due to the distribution of times. This noninteracting model, also discussed in [5], does not predict a logarithmic relaxation in $\epsilon''(\omega)$.

Our samples included a 5% potassium-doped silicate ($K_2O : SiO_2$) glass which was ground down from bulk to 20 μm thickness [11], a free standing 15 μm Mylar film, and a 3 μm thick SiO_x sample that was studied earlier by Salvino *et al.* [4]. The Mylar and potassium silicate samples had evaporated chrome-gold electrodes. The reactively sputtered SiO_x film had sputtered niobium electrodes. The techniques used are described by Salvino *et al.* [4].

In our first set of experiments we applied a large dc bias field to our samples and then measured the ac dielectric response after the application. We observed a jump in $\epsilon'(\omega)$ followed by a logarithmic relaxation of the form $\delta\epsilon' = \Delta - S_t \ln(t - t_0)$. Figure 1 shows two data sets

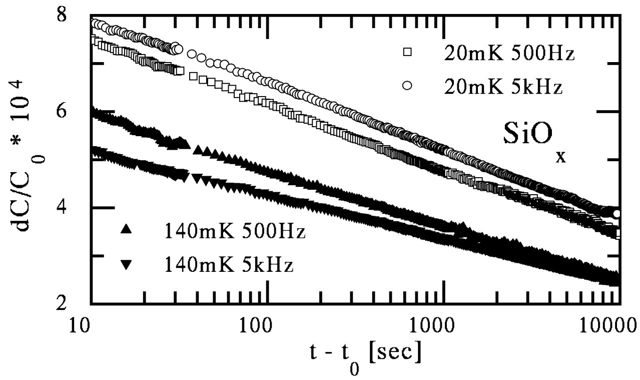

FIG. 1. Relaxations in $\epsilon'(\omega)$ of the SiO_x sample at temperatures above and below the minimum of $\epsilon'(\omega, T)$ demonstrate the frequency independence of the response at low temperatures (the two parallel lines).

from this experiment on the SiO_x sample, one at 140 mK and the other at 20 mK using both 500 Hz and 5 kHz bridge excitations. The ratio of the slopes at the two different frequencies is very close to unity at the lower temperature (the two parallel lines). At the higher temperature the ratio of the slopes is about 1.4 (the two intersecting lines). This behavior is common to all our samples, but the crossover temperature to the frequency independent regime varies from sample to sample. The ratios of the slopes and other parameters are shown in Table I. An important parameter for comparing temperature scales between samples is $T_{\min} \propto \omega^{1/3}$, the temperature of the minimum in $\epsilon'(\omega, T)$. The minimum results from a crossover between a regime where relaxation to the phonon bath dominates the TLS contribution to $\epsilon'(\omega)$ and one where resonant tunneling dominates. Considering $T/T_{\min}$ as the appropriate temperature scale (see Table I), it is not surprising that we have to go to a lower temperature for the Mylar than for the SiO_x to see the frequency independence of S_t. We observed logarithmic relaxations for up to four decades in time in all our samples including the Mylar polymer sample which is not a glass in the usual sense.

We applied dc bias fields up to 8 MV/m, although the ratio of the relaxation slopes was independent of the applied dc bias, and used ac fields ranging from 5 to 50 kV/m to measure the relaxation. The slope ratios were independent of the ac field amplitude up to the point where we started to elevate the temperature of the sample

TABLE I. The ratio of the relaxation slopes is $R_s = S_t(500\ \text{Hz})/S_t(5\ \text{kHz})$. The temperature slope below $T_{\min}$ is given as $S_T = \partial(C/C_0)/\partial \ln(T/mK)$ at 1 kHz, as for $T_{\min}$.

Sample	$T_{\min}$ (mk)	S_T	T_l (mk)	$R_s(T_l)$	T_h (mk)	$R_s(T_h)$
SiO_x	65	5.3×10^{-3}	20	1.03 ± 0.9	140	$1.48 \pm .07$
K: SiO_2	55	8.6×10^{-4}	13	$0.92 \pm .10$	140	$1.49 \pm .17$
Mylar	23	3.1×10^{-4}	10	$0.97 \pm .05$	140	$1.34 \pm .24$

($\mathbf{E}_{\rm ac} \approx 150$ kV/m) even though at this field $\epsilon'(\omega)$ is no longer field independent. The results presented are not influenced by nonlinear effects or by heating. We used a series of relaxations to determine the ratio of the slopes, and found that the scatter in the ratios cannot be explained by the uncertainty in the fits. We attribute this scatter to the slow relaxations which cause hysteresis in the samples. Hysteretic behavior in the glasses has been observed earlier [4].

We have also swept the dc field continuously using a triangular wave ($10\ \mathrm{kV\,m^{-1}\,s^{-1}}$), while measuring the capacitance. The change in $\epsilon'(\omega, \mathbf{E})$ with dc field is shown in Fig. 2 for the Mylar sample. At low temperatures one can see a sharp hole in $\epsilon'(\mathbf{E})$ in all our samples centered around zero, $\approx$1 MV/m wide, similar to those reported by Salvino *et al.* This hole broadens and becomes shallower at higher temperatures, and is almost undetectable in the sweeps at 140 mK. At 140 mK the change in $\epsilon'(\omega)$ due to the bias field is 1.4 times larger at 500 Hz than at 5 kHz, while at low temperatures the traces are almost the same. This shows that although the sharp hole in $\epsilon'(\omega)$ is not visible at 140 mK, a broad minimum remains. As discussed above, this also suggests that the change in $\epsilon'(\omega)$ due to the bias field is frequency dependent at high temperatures and not at low temperatures. This again is true for all our samples, provided the low temperature data were taken well below $T_{\rm min}$.

We always saw hysteresis in the continuous sweeps. This is due to the fact that our sweep rate was slow compared to the fast initial relaxation in $\epsilon'(\omega)$. To avoid this problem we performed a discreet sweep by rapidly applying a bias field $\mathbf{E}$ and recording the relaxation of $\epsilon'(\omega)$ for 1000 s repeatedly for different dc fields. Fitting the data at each bias point by a logarithmic relaxation, we get the parameters Δ and S_t mentioned earlier in terms of the bias field. We find the ratio between Δ and S_t to be independent of $\mathbf{E}$, so that the relaxation can be written as $\delta\epsilon'(t) = f(|E_{\rm dc}|)[\Delta - S_t \ln(t - t_0)]$. The function f is approximately linear in $E_{\rm dc}$ outside the zero bias hole and is consistent with $f \propto \ln^2(pE_{\rm dc}/k_BT)$ (where p is the average dipole moment) as predicted by the interacting model, in agreement with the continuous sweeps.

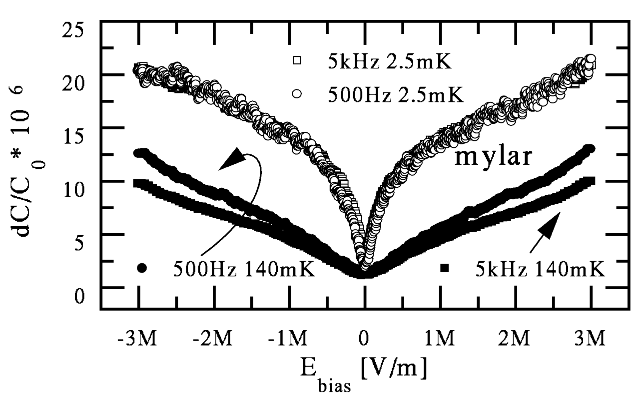

FIG. 2. dc bias sweeps on the Mylar sample showing frequency independence at low temperatures and larger response at 500 Hz above the minimum of $\epsilon'(\omega, T)$. This frequency dependence suggests that the minimum in $\epsilon'(\omega, \mathbf{E})$ is very broad.

To test the theoretical models, we have compared the measured temperature, frequency, and time dependence of the relaxation in $\epsilon'(\omega)$ and $\epsilon''(\omega)$ to the theories. The biggest discrepancy between the interacting and noninteracting models is that the noninteracting model does not predict any frequency dependence to the relaxation. This is not supported by the data; we see frequency dependent relaxation rates at high temperature for all samples. This frequency dependence vanishes at low temperatures, as predicted by the model involving the $1/r^3$ interactions. It is surprising that it is possible to describe the observed behavior well by calculating the dielectric response with a correction to the density of states due to interactions and then treat the TLS as independent. It is still possible, however, that some of the behavior observed arises from a subset of TLS which are independent, or from coupled TLS which still behave according to the Bloch equations. To distinguish further between the two models we have tested additional predictions described below.

Probing the noninteracting model we looked for a crossover from the adiabatic to the nonadiabatic regime by applying the dc bias to the Mylar and SiO_x samples with rise times ranging from 3 μs to 200 ms at low temperatures. We found the jump Δ and the slope S_t of the relaxation in $\epsilon'(\omega)$ to be independent of rise time within error bars (5%) for both limits. We also looked for a deviation from a logarithmic relaxation at large times, but could not find one up to 10^4 s, despite the fact that the rise times differed by up to 5 orders of magnitude and that the ratio of the rise time to the total measurement time was as large as 10^{10}. The lack of rise time dependence seems to exclude a spin dynamical effect, which should manifest itself as a nonmonotonic relaxation in $\epsilon'(\omega)$ after 1000 s with a rise time around 10 ms based on the prediction of the noninteracting model.

Since the noninteracting model does not predict a logarithmic decay in the dielectric dissipation, we measured long relaxations with the SiO_x sample while monitoring the change in $\tan(\phi) \propto \epsilon''(\omega)$. We observed $\delta\epsilon''(\omega)$ to decay logarithmically for more than four decades in time. The fractional change in $\epsilon''(\omega)$ due to the dc bias is much larger than the fractional change observed in $\epsilon'(\omega)$ (by approximately a factor of 100). Sweeps of the bias field while measuring dissipation show a minimum in $\epsilon''(\omega)$ centered around zero field similar to those in $\epsilon'(\omega)$. The dissipative response due to a dc bias decreases monotonically with decreasing temperature below the plateau mentioned below. The temperature dependence of this effect does not scale with the temperature dependence of the equilibrium $\epsilon''(\omega)$. Furthermore, the frequency dependence of $\delta\epsilon''(\omega, E)$ is complicated and does not scale as the equilibrium dissipation. The fact that we see a logarithmic relaxation in $\epsilon''(\omega)$ contradicts the noninteracting model and supports the interacting model that predicted this.

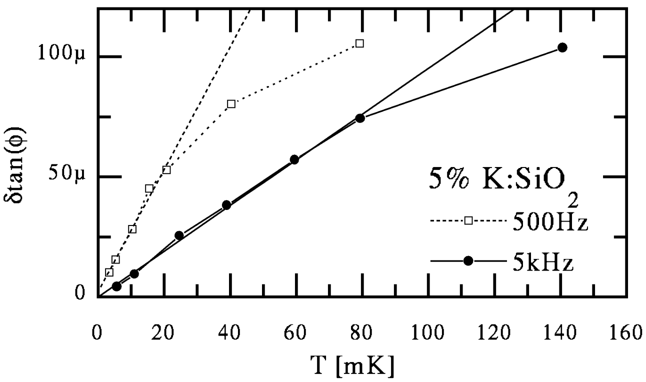

FIG. 3. The change of the loss tangent in $K:SiO_2$ for two frequencies shows the linear regime below the plateau.

Motivated by the evidence of interactions in the nonequilibrium response of the dielectrics, we looked for discrepancies between the equilibrium behavior of our samples and the standard independent TLS model. Burin and Kagan [12] suggest that a non-phonon-assisted relaxation mechanism due to $1/r^3$ interactions should contribute to the dielectric loss as $\delta\epsilon''(\omega) \propto T$. Such a term should dominate the dielectric loss at low temperatures. The change in loss at 5 kHz for the potassium-doped sample is shown in Fig. 3. The loss is linear in temperature up to 80 mK where it turns into the plateau. At 500 Hz the rise is steeper and deviates from linearity around 20 mK, while the plateau is at the same height as the 5 kHz data within experimental error as expected. This agrees qualitatively with the model [12], but the slope of the linear regime should scale like $1/\omega$ while our data suggest a weaker dependence on frequency. The Mylar and SiO_x samples show the same behavior, but the change in loss tangent of the SiO_x sample is much larger, of order 10^{-3}. One may argue that the anomalous temperature dependence of the dissipation could result from a two dimensional phonon spectrum in our thin samples, but this would lead to a $\epsilon''(\omega) \propto T^2$ behavior which is not seen. We believe there is three dimensional phonon behavior in our samples since all samples except the Mylar are bonded to a crystalline substrate and all show the same qualitative behavior, including the Mylar sample.

The lack of a T^3 dependence to $\epsilon''(\omega)$ at low temperature in the audio frequency range has been observed by other groups, for example, [13], but $\epsilon''(\omega) \propto T$ was not mentioned. The T^3 dissipation which one expects from the independent TLS model is observed in acoustic experiments using frequencies in the MHz range [14]. We regard the linear and sublinear behavior at low temperatures as an important deviation from the standard TLS model. The observations are in qualitative agreement with the interactive model by Burin and Kagan [12] which was motivated by low frequency acoustic measurements by Esquinazi, König, and Pobell [15].

In conclusion, the following observations strongly support a model involving a dipolar gap in the distribution of two level systems versus local field: frequency dependence in the relaxations of $\epsilon'(\omega)$ at high temperature and none at low temperatures, the lack of a dependence of these relaxations on the rise time of the bias field, and the logarithmic relaxation in $\epsilon''(\omega)$. Further, the importance of interactions between TLS on the equilibrium dielectric response is consistent with the anomalous behavior of the dielectric dissipation at low temperatures, $\epsilon''(\omega) \propto T$. We expect the interactions will also manifest themselves in other properties like the specific heat of disordered systems at low temperatures.

The authors wish to acknowledge useful discussions of their data and the theoretical background with Alexander Burin and with Clare Yu and Hervè Carruzzo. In addition, we wish to thank John Schroeder, who provided us with the material for the potassium-doped samples. D.N. wishes to acknowledge fellowship support from the Fannie and John Hertz Foundation. This work was supported under Department of Energy Grants No. DE-FG03-90ER45435 and No. DE-FG05-89ER75517.

[1] R. Zeller and R. Pohl, Phys. Rev. B **4**, 2029 (1971).
[2] J.C. Lasjaunias, R. Maynard, and M. Vandorpe, J. Phys. (Paris) **39**, 973 (1978).
[3] C.C. Yu and A.J. Leggett, Comments Condens. Matter Phys. **14**, 231 (1988).
[4] D.J. Salvino, S. Rogge, B. Tigner, and D.D. Osheroff, Phys. Rev. Lett. **73**, 268 (1994).
[5] A.L. Burin, J. Low Temp. Phys. **100**, 309 (1995).
[6] S. Kirkpatrick and C.M. Varma, Solid State Commun. **25**, 821 (1978).
[7] P.J. Anthony and A.C. Anderson, Phys. Rev. B **20**, 763 (1979).
[8] H.M. Carruzzo, E.R. Grannan, and C.C. Yu, Phys. Rev. B **50**, 6685 (1994).
[9] P.W. Fenimore and M.B. Weissman, J. Appl. Phys. **76**, 6192 (1994).
[10] A. Würger, Z. Phys. B **93**, 109 (1993).
[11] W.M. MacDonald, A.C. Anderson, and J. Schroeder, Phys. Rev. B **31**, 1090 (1985).
[12] A.L. Burin and Y. Kagan, JETP Lett. **80**, 761 (1995).
[13] J. Classen *et al.*, Ann. Phys. (Leipzig) **3**, 315 (1994).
[14] S. Hunklinger, in *Proceedings of the International School on condensed Matter Physics, Varna, 1988,* edited by M. Borissov, N. Kirov, and A. Vavrek (World Scientific, Singapore, 1989), p. 113.
[15] P. Esquinazi, R. König, and F. Pobell, Z. Phys. B **87**, 305 (1992).