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F. Ladieu,[1,*] J. Le Cochec,[1] P. Pari,[2] P. Trouslard,[3] and P. Ailloud[1]
[1]*DSM/DRECAM/LPS, C. E. Saclay, 91191 Gif-sur-Yvette Cedex, France*
[2]*SPEC, C. E. Saclay, 91191 Gif-sur-Yvette Cedex, France*
[3]*INSTN/LVDG, C. E. Saclay, 91191 Gif-sur-Yvette Cedex, France*
(Received 25 September 2002; published 19 May 2003)

The 1 kHz real part χ' of the dielectric constant of a structural glass (a-$SiO_{2+x}C_{1+y}H_z$) was measured at low temperature T. Reducing the sample thickness h below 100 nm weakens the slope $|\partial\chi'/\partial T|$ for $T \lesssim 0.1$ K, for all measuring fields E. This contrasts with the predictions of the two-level system (TLS) model but is in agreement with the recently proposed delocalization of excitations derived from a field-induced TLS-TLS interaction mechanism. For small h this interaction is screened, which explains the h effects on χ'. Hence, interactions must play a key role in standard thick samples as soon as $T \lesssim 0.1$ K.

DOI: 10.1103/PhysRevLett.90.205501 PACS numbers: 61.43.Fs, 72.20.Ht, 72.20.My, 77.22.Gm

For more than 30 years, amorphous solids have been widely investigated at low temperature T [1]. Their properties turn out to be both "universal" (i.e., quasi-independent from their chemical composition) and strongly different from their crystalline counterparts. This is explained within the tunneling two-level system (TLS) model [2] where some atomic systems fluctuate between two neighboring energy minima separated by a potential barrier, which, at low T, is crossed by tunneling.

Even if the low density of glasses (in comparison with their crystalline counterpart) may justify [3] the existence of "voids" and thus the TLS framework, the ability of the standard noninteracting TLS model to account for experiments is very intriguing. Indeed, by extracting from measurements the coupling strength of a TLS to phonons, one finds [4] that TLS's are strongly coupled via virtual phonons. A tunnel transition on a given TLS deforms elastically the neighboring matrix, thus inducing an energy change $U \propto 1/r^3$ for a TLS located at a distance r. For two neighboring TLS's [5] one gets $U(r = 1\ \text{nm}) \simeq 10$ K. Similarly, since many TLS's are charged, they interact through dipolar interaction, mediated by virtual photons, giving an interaction energy [5] in the same range of 10 K between neighboring TLS's. This large energy scale contrasts with the (very few) experimental results that suggest weak interactions between TLS's. In the 100 mK range, only the small nonequilibrium effects of the kHz dielectric susceptibility χ [6], as well as its small unexpected H dependence [7], have been related to interactions. In the mK range, somewhat larger effects, such as the ultralow T plateau [8] of the dielectric constant, and the internal friction behavior [9], were ascribed to TLS interactions.

This work provides strong evidence of the key role played by TLS's interactions in the 100 mK range. We show that reducing the thickness h of our glassy samples below 100 nm strongly affects the real part χ' of the 1 kHz dielectric constant. The $\chi'(T)$ minimum, which occurs at a given T_{rev} for $h > 100$ nm, progressively shifts to lower T as h is decreased (and finally disappears for $h \simeq 12$ nm). We show that this behavior contradicts the noninteracting TLS model and is most likely related to TLS interactions. Many theories dealing with interactions in the TLS model have been proposed. In Ref. [10], it is suggested that interactions are renormalized to zero by frustration, whereas in Ref. [11] interactions are assumed to supersede disorder. More recently Burin *et al.* [12] proposed a mechanism that accounts for the reported data, once properly factored in the numerical determination of χ' [13].

The samples were all produced similarly: on a vitreous a-SiO_2 0.1 mm thick substrate, a layered Cu/glass/Cu/glass structure was deposited, by using a multichamber system excluding exposure to air during the whole process [14]. The 15 nm thick Cu electrodes were evaporated (at 0.1 nm/s). The glass layers of a-$SiO_{2+x}C_{1+y}H_z$ (with $x \simeq 2.9$, $y \simeq 0.9$, $z \simeq 1.2$) were made from TetraEthylOrthoSilane with a 13 MHz vapor plasma, and x-ray analysis confirmed their amorphous nature [15]. The autopolarization was set to -100 V, the incoming flux to 2 sccm, and the pressure to 0.80 Pa. The resulting glass deposition rate was 0.1 nm/s, allowing one to select the thickness h of the internal glass layer by choosing the deposition time. Since the value of h is crucial, it was independently measured by three other methods that all gave compatible results: (i) *in situ* laser interferometry was realized onto a Si substrate placed close to the sample during the glass deposition; (ii) the glass layer grown onto the Si substrate was irradiated by a deuteron 0.91 MeV beam allowing, using the nuclear reaction on $_{16}O$, an h estimate; (iii) the value of the capacitance $C \propto \chi'$ was checked to scale with the expected h. The 15 nm thick top glass layer hinders any spurious atmospheric effect during the cryogenic experiment. The sample was glued inside a copper box connected to the mixing chamber of the $_3He/_4He$ dilution

refrigerator. Semirigid coaxial wiring was ensured from the cold copper box up to the 2500-Andeen capacitance bridge. A capacitance c_F, twice as large as that of the sample, was set in parallel of each cable, so as to filter high frequency parasitic fields. The data remained unchanged when c_F was halved, proving the filtering efficiency.

The $\chi'(T, E)$ behavior of six samples, with h ranging from 12 to 800 nm, is reported in Figs. 1–3, where $\delta C(T) = C(T) - C(0.5\ \mathrm{K})$ and $\delta C_{\max} = C(0.5\ \mathrm{K}) - C_{\min}$, while $C_{\min}$ is the minimum value of C in our T experimental range (14 mK; 0.5 K). Figure 1 shows that, for a given measuring field $E = 40$ kV/m, the temperature T_{rev}, where $\chi'(T)$ is minimum, *gradually shifts towards lower values as h is decreased.* For the thinnest sample, the minimum is suppressed for fields lower than 250 kV/m. At $E = 40$ kV/m, the simplest way to describe the $\delta C(T)$ behavior of the thinnest sample is $\delta C(T) \propto T^{\alpha}$, where α is equal to 1.35 ± 0.15 above 70 mK and to 2.75 ± 0.4 below. This strongly contrasts with the usual logarithmic behavior $\delta C(T) \propto \pm \ln T$, valid for thicker samples as well as for all previously studied glasses [16]. Figure 2 shows that the lowering of T_{rev} when decreasing h occurs for all E (except for $E \geq 1$ MV/m where some heating arises; see below). However, this decrease occurs only for $h \lesssim 100$ nm since the 200 and 800 nm samples behave similarly. Finally, Fig. 3 shows, for the smallest h, that a minimum of $\chi'(T)$ is recovered when $E \gtrsim 1$ MV/m, but the value of the slope $|\partial\chi'/\partial T|$ below T_{rev} is smaller than that of thick samples.

The above properties of $\chi'(T, E, h)$ are intrinsic to the glassy state. Indeed, as we show below, they come neither from a heating effect nor from a variation of the glass composition with h:

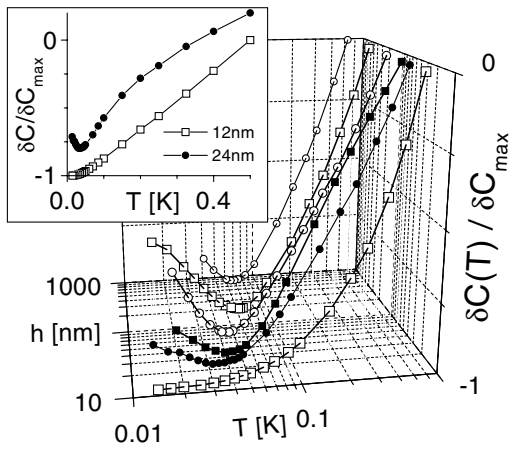

FIG. 1. Relative variation with T of the capacitance, for a given measuring field $E = 40$ kV/m. As the sample thickness h decreases the minimum shifts towards lower T and disappears for the smallest h. This contradicts the standard *noninteracting* TLS model. Inset: same plot (T in linear scale); the 24 nm curve is vertically moved (+ 0.2) for clarity. Note that for $h = 12$ nm, the standard $\ln T$ behavior is not obeyed.

(i) The E-induced dissipated power $\mathcal{P}$ might heat the sample to a temperature above the measured T. This explanation is ruled out since the data of Fig. 1 are taken at constant E. Note that one has $\mathcal{P} \propto E^2 h$; i.e., $\mathcal{P}$ increases with h at a given E. Thus, if the measurements were affected by heating, the observed trend would be *the opposite* of that of Figs. 1 and 2. The presence of heating can be suspected only for the two thickest samples and for $E \gtrsim 1$ MV/m, where T_{rev} is slightly lower at $h \geq 200$ nm than at $h = 75$ nm. To check this point, the thermal resistance $\mathcal{R}$ between the sample and the cryostat was estimated as follows: first for $E > 10$ kV/m, the law $T_{\mathrm{rev}} \propto E^{1/2}$ was assumed to hold (see below) for a perfectly well thermalized sample; second, the (small) difference between this law and the measured T_{rev} in the range 10–300 kV/m was attributed to a (small) heating effect. From the two thickest samples one gets $\mathcal{R} \simeq 50$ MK/W at 100 mK: this is the good order of magnitude to account for the above-mentioned behavior of T_{rev} when $E \gtrsim 1$ MV/m and $h \geq 200$ nm. Finally, one finds $\mathcal{R} \propto T^{-3}$, as expected for boundary Kapitza resistances.

(ii) Besides, one may imagine that the effect of Figs. 1–3 comes from the fact that the glass composition depends on h, even for a given set of plasma parameters. The components of the plasma may be mixed with the Cu of the first electrode and yield a "boundary" layer, of thickness b_0, with a chemical composition that strongly differs from the rest of the "normal" glass (of thickness $h - b_0$). Since the value of T_{rev} slightly depends on the glass composition [16], one may wonder if the data could be explained by combining the susceptibilities of these two dielectrics in series. This is not the case. Indeed, taking for the boundary layer the 12 nm sample, and for the normal glass the 800 nm sample, one finds, for the 22 nm thick sample, that $\delta C(T)$ should, by far, be dominated by

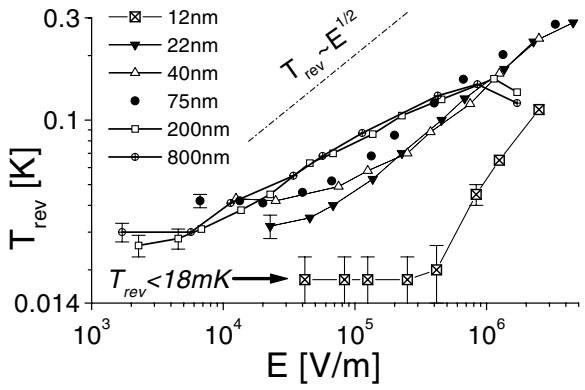

FIG. 2. Temperature T_{rev} of the minimum of $\chi'(T) \propto C(T)$, as a function of E, for various h (labeling the curves). The main point is that for $E \geq 10$ kV/m, T_{rev} *decreases with h* (except for $E \gtrsim 1$ MV/m; see text). The minimum of χ' even disappears for $h = 12$ nm and for $E \leq 250$ kV/m, which is indicated by "$T_{\mathrm{rev}} < 18$ mK." For clarity, the ± 4 mK error bar is shown only for a few lowest T_{rev}.

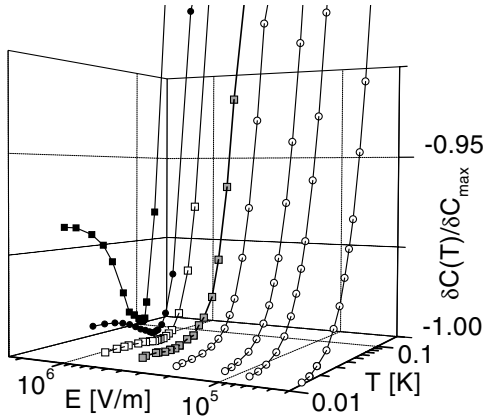

FIG. 3. Relative variation of the capacitance with T, for various E and for $h = 12$ nm. No minimum occurs in $\chi'(T)$ down to 14 mK for the four smallest E's, but a minimum reappears for larger E's. At $E \simeq 0.4$ MV/m (gray symbols) one may have $T_{\rm rev} = 19$ mK. The vertical scale is reduced compared to that of Fig. 1, since the variation of $\chi'(T < T_{\rm rev})$ is weaker than in thick samples.

the "thin" T^{α} behavior, for any E. This is clearly contradicted by Fig. 1 where the standard $\ln T$ trend holds for the 22 nm thick sample. Even if the 12 nm thick sample is discarded because of its unusual T behavior, the scenario of two dielectrics in series can be further tested by using the 22 nm sample as the new "boundary layer." For any given E, the resulting $T_{\rm rev}$ for the 75 nm thick sample should be only 5 mK below that of the thickest sample. This is clearly contradicted by the data of the 100 kV/m range of Fig. 2.

At this step, it is clear that decreasing h below 100 nm qualitatively changes the physics of the glass sample. This contradicts the standard (i.e., noninteracting) TLS picture. This model accounts for the decrease of χ' above $T_{\rm rev}$ by the progressive freezing of the diagonal (or relaxational) part χ'_z of the susceptibility, whereas the increase of χ' below $T_{\rm rev}$ comes from the off-diagonal (or resonant) part χ'_x (see Ref. [17]). Because of its purely quantum nature, χ'_x grows when T decreases, as do all quantum effects. In other words, according to the standard model, χ' is dominated by the dynamics of individual TLS's (see Ref. [18]). Since the TLS's size is below 1 nm, no h effect can be explained in the range of tens of nm, contrary to the data reported above.

This failure of the standard TLS model strongly contrasts with its former success, in particular, its ability to account for the $\chi' \propto \pm \ln T$ behavior as well as for the order of magnitude of $T_{\rm rev}$ [1,16]. In fact, it has been very recently discovered that the agreement holds only in the "linear regime" (i.e., small E's) where χ', and thus $T_{\rm rev}$, do not depend on E. Indeed, as shown in Ref. [13], the standard TLS model does not account for the nonlinear behavior of $\chi'(T, E)$ (such as that reported in Ref. [16] and that of our $h > 100$ nm samples). Indeed, due to the quantum nature of χ' below $T_{\rm rev}$, χ' is strongly *depressed* by a strong measuring electric field E at a given T. This is due to the quantum saturation phenomenon coming from the fact that increasing E decreases the population difference between the two energy levels. As the Rabi oscillations produced by E on the upper level are in phase opposition with respect to those produced on the ground level, the quantum response, once averaged on many independent TLS's, tends to zero when E is increased. This was checked by solving numerically the Bloch equations of TLS's [13] with a nonperturbative method (see the dotted lines in Fig. 4).

Both the nonlinear measurements and the h effect on χ' can be accounted for by using the field-induced TLS-TLS interaction proposed by Burin *et al.* [12]. The idea is to consider a thermally excited TLS of gap ϵ and to seek for the probability ρ that such an excitation delocalizes by hopping to another TLS of gap ϵ' located at a distance λ from the first one. The probability ρ is large only if $|\epsilon - \epsilon'| \leq U(\lambda)$, where $U(\lambda)$ is the interaction coupling the two TLS's. With the usual density $\bar{P}$(independent on ϵ) of gaps in glasses, one finds that ρ is extremely small at $E = 0$, but that a finite E strongly favors the delocalization of excitations. This is due to the fact that E modulates any ϵ by an amount $e_{\rm dip} = pE/\epsilon_r$, where $p \simeq 1$ D is the TLS dipole and $\epsilon_r \simeq 5$ is the glass dielectric constant [6,16]. Thus, all the TLS's whose gaps differ from each

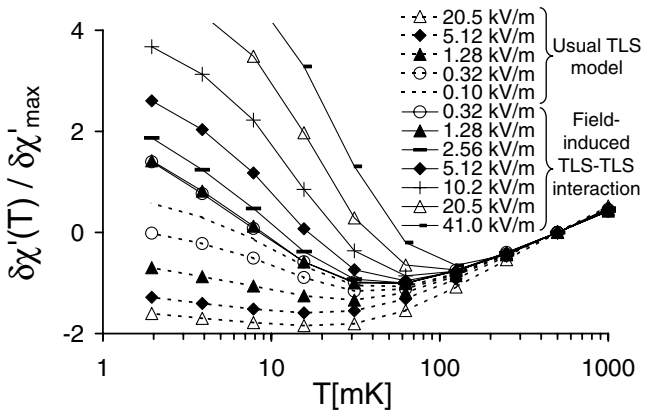

FIG. 4. $\chi'(T, E)$ was computed in Ref. [13] by solving Bloch equations. The *noninteracting* TLS model is shown in dotted lines: due to the quantum saturation effect, $\chi' \propto C$ is strongly depressed when raising E at low T. This suppresses the minimum of $\chi'(T)$ for experimental fields $E > 40$ kV/m and could model the 12 nm sample where TLS's interactions are *strongly reduced.* Including TLS's interactions through the E-induced mechanism of Ref. [12] restores the χ' increase with E (at any given T), as well as that of $T_{\rm rev}$. This models the usual $\chi'(T, E)$ of thick samples. Finally, since strong E's decrease the distance λ between interacting TLS's, one recovers the usual behavior of $\chi'(T)$ when $\lambda(E) < h$. This explains, for $h = 12$ nm, the appearance of a $\chi'(T)$ minimum for strong enough E, as in Figs. 2 and 3.

other by less than e_{dip} form an "infinite cluster" within which the delocalization of excitations occurs.

As far as the nonlinear behavior of χ' is concerned, this mechanism leads to the following predictions: (i) Energy diffusion enhances the diagonal part χ'_z of the susceptibility, by decreasing the associated relaxation time τ_1. This is of great qualitative interest since, as stated above, χ'_x is mainly depressed by E and thus cannot account for the increase of χ' with E at low T. (ii) The rate of delocalization (i.e., $1/\tau_1$) increases with E, and the associated increase of χ'_z with E overcomes the E-induced depression of χ'_x. (iii) The rate $1/\tau_1$ grows when T decreases, since quantum coherence is needed for the delocalization of excitations. Figure 4, drawn from Ref. [13], shows that the mechanism of Burin *et al.* allows one to recover a nonlinear $\chi'(T, E)$ behavior in qualitative agreement with that of our two thickest samples (or that of Ref. [16]). Along the same lines [13], one predicts $T_{\mathrm{rev}} \propto \sqrt{E}$, as seen in Fig. 2 in the nonlinear regime ($E \geq 10$ kV/m).

The delocalization of excitations also accounts for the thickness effects reported here. Indeed, dipolar interactions are strongly screened by the electrodes [19], i.e., by the fact that their numerous electrons intercept and cancel the electric field responsible for the interaction between TLS's. This weakens $U(\lambda)$ for distances $\lambda \gtrsim h$. Since the distance $\lambda = (\bar{P} e_{\mathrm{dip}})^{-1/3}$ between two neighboring TLS's of the infinite cluster is typically 60 nm for $E =$ 40 kV/m, the mechanism of Burin *et al.* is expected to be progressively switched off as h decreases in this range. Since this mechanism is responsible for the behavior of $\chi'(T \leq T_{\mathrm{rev}})$, one understands that T_{rev} should decrease with h, as seen on Fig. 2. As an extreme consequence, just as in Fig. 1, T_{rev} could even disappear for the thinnest sample due to the complete suppression of the delocalization of excitations (and to the E-induced depression of χ'_x valid at any h).

Besides, due to the E-induced modulation of ϵ, in the scenario of Burin *et al.*, λ decreases for strong E, opening the possibility to recover $\lambda < h$ for large enough E. This could account for Fig. 3 where, even for $h = 12$ nm, one recovers a minimum for $\chi'(T)$.

In conclusion, decreasing the thickness of glass samples below 100 nm changes the physics of the real part of the kHz dielectric susceptibility. This experimental result strongly supports the key role of dipole-dipole interactions up to 100 mK. The main features of the reported data can be accounted for by assuming that TLS's interactions occur via an E-induced delocalization of excitations between quasisimilar TLS's.

We thank M. Ocio and E. Vincent for lending us their cryostat and L. Le Pape for experimental support. We especially thank J.-Y. Prieur and J. Joffrin (CNRS, Orsay) for very encouraging scientific discussions, D. L'Hôte and A. Forget for experimental help, and J. P. Bouchaud for helping us to write this Letter.

*Electronic address: ladieu@drecam.cea.fr

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[15] X-ray experiments (wavelength 0.154 nm), carried out onto layers deposited on Si-111 substrates, give *the two main features of amorphous solids:* (i) the layers do not yield any Bragg peak; (ii) setting the angle of incidence $\theta = 5°$ and scanning the detector angle, each layer gives a "flattened hump" ranging from 15° to 35°. This flattened hump is mainly *unchanged* when h ranges from 40 nm to 1 μm. Since the "hump's height/noise" ratio worsens at small h, we cannot exclude that this "hump" is somehow deformed at small h. Should this be the case, it *might* lead to a small variation of the elementary dipole p_0 (or of the TLS density of states $\bar{P}$) with h. This is of no consequence here since, in the standard noninteracting TLS model, $\delta C/\delta C_{\mathrm{max}}$ as well as T_{rev} depends neither on $\bar{P}$ nor on p_0.
[16] S. Rogge *et al.*, Phys. Rev. B **55**, 11 256 (1997).
[17] The susceptibility χ' is given by $\chi' = \chi'_z + \chi'_x$; see Ref. [13] for a detailed explanation.
[18] More precisely, in the standard TLS model, TLS-TLS interactions are assumed to be so small that they enter only in the phase coherence time τ_2. Typically $\tau_2 \simeq$ 10 μs for $T = 20$ mK; i.e., the associated energy $\propto \tau_2^{-1}$ is 1 μK, which is much smaller than any relevant energy scale. That is why, according to the standard model, "χ' depends only on what happens on the scale of individual TLS's."
[19] The distance λ between the two TLS's turns from $\lambda \propto (e_{\mathrm{dip}})^{-1/3}$, as derived in the text for $\lambda < h$, to $\lambda \propto (e_{\mathrm{dip}})^{-1/2}$ when $\lambda > h$. This "simple geometric effect" somewhat enlarges λ and thus reduces both the elastic and the dipolar parts of U. It is not easy to evaluate the importance of the "geometric effect" with respect to that of screening by the electrodes. Whatever it may be, this does not change our conclusion that the h effects reported here come from a reduction of TLS-TLS interactions.