

# Effects of Large Nuclear Quadrupoles on Dielectric Properties of Glasses at Very Low Temperatures

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## Abstract.

The universal behaviour of amorphous solids at low temperatures, governed by atomic tunneling systems as described by the standard tunneling model, has long been a generally accepted fact. In the last years, however, measurements of dielectric two-pulse polarization echoes have revealed that nuclear quadrupole moments involved in atomic tunneling systems can cause specific material-dependent effects in magnetic fields. We have performed measurements of the dielectric properties of the two multicomponent glasses N-KZFS11 and HY-1, containing several percent of tantalum oxide and holmium oxide respectively. As <sup>181</sup>Ta and <sup>165</sup>Ho both carry very large nuclear quadrupole moments, these glasses are ideal candidates to study the influence of nuclear quadrupole moments on the properties of glasses at very low temperatures. Our measurements not only show unique dielectric behaviour in both glasses, but also differ significantly from various predictions of the standard tunneling model.

## 1. Introduction

The low temperature properties of amorphous solids are governed by atomic tunneling systems. A widely accepted phenomenological description of these tunneling systems is given by the standard tunneling model (STM) [1, 2]. The model assumes that the tunneling systems can be described as particles moving in double-well potentials. The tunneling motion results in a ground state splitting of the double-well potential given by energy splitting  $E = \sqrt{\Delta_0^2 + \Delta^2}$ , where  $\Delta_0 = \hbar\Omega e^{-\lambda}$  denotes the tunneling splitting determined by the ground state energy of the double-well potential  $\hbar\Omega$  and the tunneling probability  $\lambda$ . The asymmetry energy  $\Delta$  reflects a possible difference in depth of the two wells. In amorphous solids the parameters are broadly distributed and a central assumption of the STM is a uniform distribution of the form  $P(\Delta, \lambda)d\Delta d\lambda = \bar{P}d\Delta d\lambda$ .

For the real part of the low temperature and low frequency dielectric susceptibility of insulating glasses, the STM predicts a minimum in the dielectric constant as a function of temperature. Below this minimum, the temperature dependence of the dielectric constants is governed by a resonant interaction of the atomic tunneling systems with the electric field, leading to a logarithmic variation  $\delta\epsilon/\epsilon = -C \ln(T/T_0)$ , where  $C$  is the coupled density of states and  $T_0$  an arbitrarily chosen reference temperature. Above the minimum, one phonon relaxational processes add to the temperature variation of the dielectric constant and lead to a logarithmic increase given by  $\delta\epsilon/\epsilon = C/2 \ln(T/T_0)$ , thus the slope ratio in the two logarithmic regimes should be  $-2 : 1$  according to the STM. The relaxational processes also lead to a frequency



dependence of the minimum temperature, which is expected to vary as  $T_{\min} \propto f^{1/3}$ , where  $f$  denotes the frequencies (for a review see for example [3, 4]).

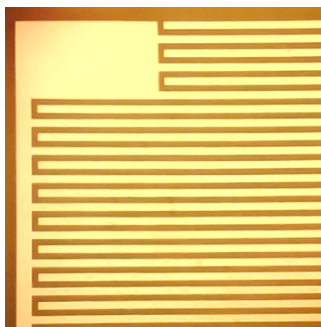
In many aspects the STM describes the thermal, acoustic and dielectric properties of glasses at low temperatures quite well. However, at the same time significant deviations have been reported that clearly show this model is incomplete. Examples are the systematic deviations from the predicted slope ratio of  $-2 : 1$  both of the dielectric constant [5, 6, 7, 8] and the sound velocity [9, 10]. The degree of observed universality has also lead to proposed modifications of the STM [11, 12].

In some glasses one also observes an unexpected dependence of the dielectric constant on small magnetic fields [13, 14, 15]. Two models have been proposed to explain the magnetic field sensitivity of the low frequency dielectric properties by a coupling of the magnetic field to loop-like tunneling paths of mutually interacting charged tunneling particles [16, 17]. The observation of a magnetic field dependence in dielectric polarization echoes performed on multicomponent glasses at about 1 GHz [18] and the explanation of such an effect as being caused by tunneling particles carrying a nuclear quadrupole moment [17, 19, 20] has naturally brought up the question whether the deviations at low frequencies are also caused by nuclear moments. Several extensions of the STM regarding the role of nuclear quadrupoles have been proposed, in which corrections for the dielectric permittivity in the resonant regime at temperatures where the thermal energy becomes comparable to the quadrupole splitting were predicted [21, 22, 23, 24]. In particular, it was argued that the nuclear quadrupole interaction of atomic tunneling atoms increases the number of levels and thereby increases the transition probability, which in turn enhances the dielectric constant. In addition, a transition from coherent to incoherent tunneling was proposed for tunneling systems whose bare tunneling splitting matches the quadrupole splitting. Since the typical quadrupole splittings of the constituents in most glasses are of the order of 1 mK and below, such effects should be only relevant at ultralow temperatures.

In order to investigate the importance of nuclear quadrupoles at low frequencies, we have conducted measurements on both a standard probe of fused silica, Herafil (Heraeus Quartz Glass), and the two multicomponent glasses N-KZFS11 (Schott) and HY-1 (Hoya) containing several percent of tantalum oxide and holmium oxide respectively. Since both elements carry very large quadrupole moments, a possible effect of nuclear quadrupoles on the dielectric constant at low frequencies should be most pronounced.

## 2. Experimental Technique

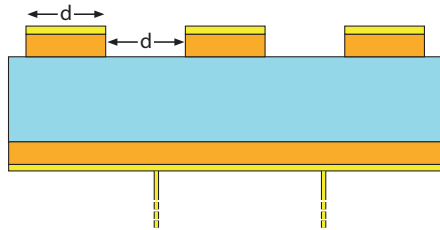
In order to enable strong thermal coupling of our glass samples to the mixing chamber of our dilution refrigerator, we used a novel capacitor design instead of a classical parallel plate setup. In our case, the capacitor consists of a thin glass plate (12 mm  $\times$  12 mm  $\times$  0.5 mm) with



**Figure 1.** Photographic image of a 1mm $\times$ 1mm section of our microstructured electrodes on one of our samples. Both the width and the pitch of the digits are 25  $\mu$ m.

microstructured interdigital electrodes on one side, as depicted in figure 1, and a metallic film as the ground plate on the other side. The metallization layers on both sides consist of a layer of

copper covered by a thin gold layer in order to prevent a degradation of the metal by oxidation, as shown schematically in figure 2. The digits have a length of about 1 cm and both a width



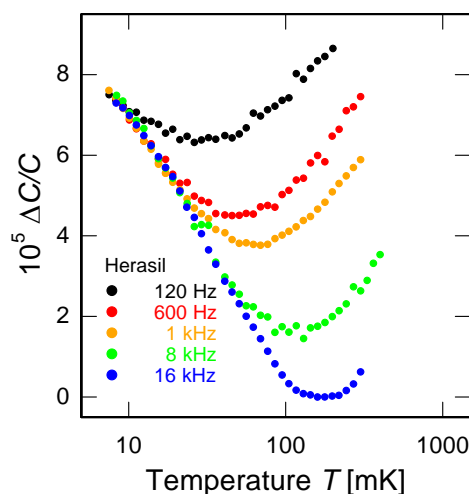
**Figure 2.** Schematic image of the capacitor with the microfabricated interdigital electrodes and the ground plate consisting of copper and gold layers on the glass sample. Both the width and the pitch of the digits are  $d = 25 \mu\text{m}$ . Thermal contact can be improved by adding gold bonds.

and a pitch of  $d = 25 \mu\text{m}$ . This very compact design has two advantages: it provides rather large absolute values of the capacitances ranging from 35 to 80 pF, depending on the dielectric susceptibility  $\epsilon$  of the samples, which makes the setup somewhat less sensitive to changes of the cable capacitance or possible stray capacitances in our experiment. At the same time our setup allows for good thermal coupling of the sample via gold bonds between the ground plate and the sample holder. The electrical contacts to the interdigital electrodes were made by aluminum bonds to minimize parasitic heating of the samples through the coaxial cables that connected the experimental stage to the room temperature electronics.

The temperature dependence of the capacitance of our samples was measured at frequencies ranging from 120 Hz to 20 kHz using a commercial capacitance bridge (Andeen Hagerling AH2700A). The excitation voltage of the bridge was chosen sufficiently low in order not to exceed the range of linear response of the samples. This was studied in detail on the Herasil sample by applying voltages both high enough to cause nonlinear responses and low enough to see a voltage independent response of the sample.

### 3. Experimental Results

The measurements on the Herasil sample yielded results similar to those seen in previous investigations on quartz glass samples [6, 7] which proved the proper functionality of our new setup. In particular, it demonstrates that nonlinear effects due to the inhomogeneous electric fields caused by the interdigital structure are negligible, if the excitation voltage is chosen sufficiently low. In figure 3, the Herasil data are shown for frequencies between 120 Hz and 16 kHz. Note that the data points for different frequencies were shifted to be aligned at the

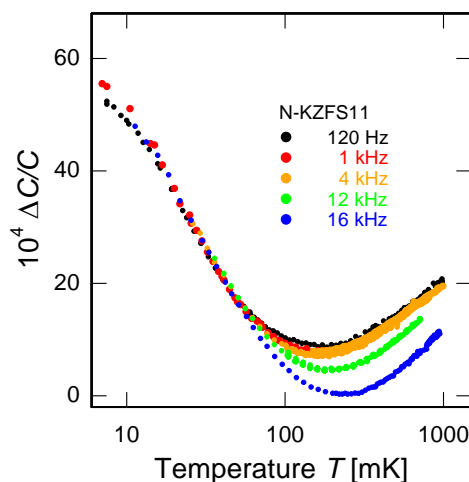


**Figure 3.** Dielectric constant of the quartz glass Herasil as a function of temperature at frequencies between 120 Hz and 16 kHz.

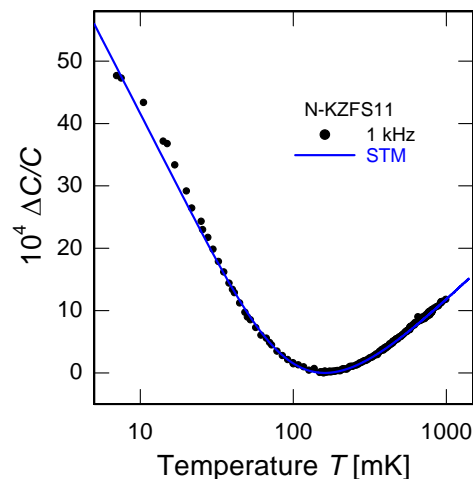
lowest temperatures, where the frequency independent resonant contribution should dominate.

It thus becomes clearly visible that all curves show the same temperature dependence in the resonant regime, while showing a clear frequency dependence at higher temperatures, where this is expected due to the relaxational contribution. Both logarithmic regimes are visible and the minimum of the dielectric constant shifts as expected for the one phonon relaxation. The data also show the typical deviation from the STM, namely the slope ratio being  $-1:1$  rather than  $-2:1$ . We would like to point out that this is an observation that has been made virtually in all low frequency dielectric measurements on glasses so far.

Let us now turn to the results obtained for N-KZFS11, containing several percent of  $^{181}\text{Ta}$ , which are shown in figure 4. As in the case of Herasil the data were shifted to align at the low temperature resonant regime. At first glance the overall behaviour looks very similar to that of the Herasil sample. One finds a minimum and both logarithmic regimes, as theoretically predicted. However, closer inspection reveals two striking differences, namely the fact that for all frequencies below 4 kHz only a very small frequency dependent shift of the minimum is seen and that the slope ratio is  $-2:1$ , as predicted by the STM but not observed in other glasses. The second remarkable observation is made clearly visible in figure 5, where the data taken at



**Figure 4.** Dielectric constant of the multi-component glass N-KZFS11 as a function of temperature at frequencies between 120 Hz and 16 kHz.

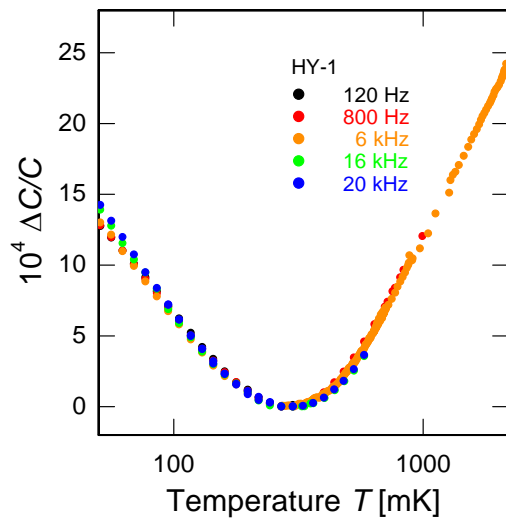


**Figure 5.** Dielectric constant of the multi-component glass N-KZFS11 as a function of temperature at a frequency of 1 kHz (●). The solid line represents a numerical fit of the data based on the STM.

1 kHz are shown together with a numerical calculation based on the STM. Obviously, for this glass the data are in very good agreement with the prediction of the STM. We would like to point out, however, that there is still a small but systematic deviation between fit and data at temperatures below about 50 mK. It is also remarkable that the temperature at which the minimum appears is significantly higher than in other samples.

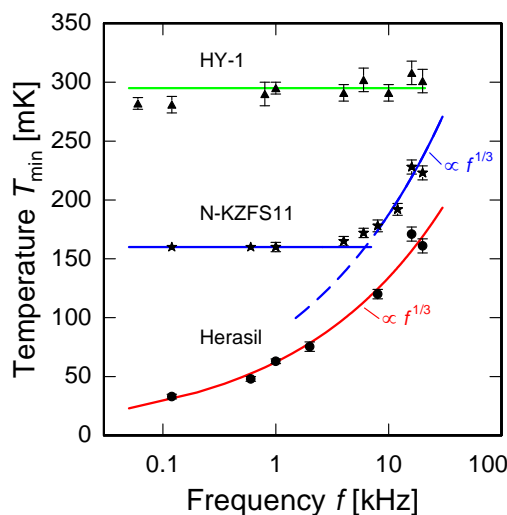
Figure 6 shows the results we have obtained for the third glass that we investigated with our new setup, the multicomponent glass HY-1 containing  $^{165}\text{Ho}$ . The measurements were performed at frequencies between 120 Hz and 20 kHz. The most striking observation for this sample is that there appears to be almost no frequency dependence of the data at all. In particular, we do not see any shift of the minimum of the dielectric constant in the entire frequency range investigated. This is a behaviour not seen at any other amorphous material before. The slope ratio, in contrast, being roughly  $-0.8:1$ , is in line with the vast majority of measurements on glasses, except the one shown above on N-KZFS11.

In figure 7 we summarize our findings regarding the frequency dependence of the minima



**Figure 6.** Dielectric constant of the multicomponent glass HY-1 as a function of temperature at frequencies between 120 Hz and 20 kHz.

for all three samples. Starting with Herasil we see that the minima appear at relatively low temperatures, indicating a strong coupling between the tunneling systems and thermal phonons. The minima shift proportional to  $f^{1/3}$ , as expected by the STM and indicated by the red solid line. The N-KZFS11 sample shows minima at systematically higher temperatures than Herasil. The temperature dependence of the minima only agrees with the STM prediction above 4 kHz and becomes frequency independent below this frequency. The minima observed for the  $^{165}\text{Ho}$  containing HY-1 glass lie at temperatures well above those of both other samples and show only a very slight frequency dependence. Although we have no detailed theoretical explanation for



**Figure 7.** Frequency dependence of the minima of all three glasses being investigated in this work. Both the frequency independent regions and the regions where the minima vary proportional to  $f^{1/3}$  are indicated by solid lines.

our findings at this point, it seems to be clear that the presence of nuclear quadrupole moments influences the low frequency dielectric behaviour of glasses quite dramatically. In particular, one is tempted to conclude from figure 7 that there is another relaxational mechanism acting besides the one phonon process in the two glasses containing elements with large nuclear quadrupole moments. We can only speculate at this point whether this relaxational mechanism is perhaps driven by the interaction of atomic tunneling systems carrying nuclear quadrupoles with all other surrounding nuclear moments. On the contrary, we have not seen any clear and systematic indications of the resonant part being modified by the presence of the quadrupole moments as suggested by [21, 22, 23, 24]. It is well possible that the temperatures we have reached in

our experiments were not sufficiently low to observe the transition from coherent to incoherent tunneling. The only hint that the resonant part is perhaps affected by presence of large nuclear quadrupole moments is the observation, that the slope ratio is different for the N-KZFS11 sample compared to all other glasses investigated so far. One could interpret the steeper resonant part seen in this experiment as the expected enhanced resonant susceptibility, however, one would then expect a similar effect for the HY-1 glass, which is not observed.

#### 4. Conclusions

We have investigated the frequency and temperature dependence of the dielectric constant of three different glasses at low temperatures using a novel setup. Two of those glasses contained elements with large quadrupole moments on the percent level and were chosen specifically to investigate a possible influence of these nuclear quadrupoles on the dielectric properties of glasses in this frequency and temperature range. Our findings clearly show that the dielectric properties of insulating glasses are not universal as predicted by the STM and that elements with large nuclear quadrupole moments strongly influence their low frequency dielectric behaviour. The striking dependence on specific elements may guide the way to solve several puzzles regarding the low temperature properties of glasses in the future.

#### 5. Acknowledgements

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