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DIRECT EXPERIMENTAL OBSERVATION OF SPECTRAL DIFFUSION IN VITREOUS SILICA AT LOW TEMPERATURES

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Résumé.— Nous présentons des expériences de "hole burning" acoustique dans de la silice vitreuse à 0,5 K. Une forte dépendance en temps est observée et est interprétée en termes de diffusion spectrale.

Abstract.— We have carried out acoustic "hole burning" experiments in vitreous silica at 0.5 K. A strong time dependence was observed and is interpreted in terms of spectral diffusion.

The anomalous thermal /1/ and acoustic /2/ properties of glasses at low temperatures can be explained by the existence of two-level systems (2LS). These states are assumed to arise from the configurational disorder present in the network of amorphous materials. In a more specific model /3/ the 2LS are ascribed to particles (of still unknown nature, but probably groups of atoms) moving in double well potentials. Transitions from one well to the other are possible even at the lowest temperatures via tunneling. The parameters characterizing such double well potentials exhibit — within certain limits — a random distribution of their values caused by the randomness of the amorphous network. The result is a broad, approximately constant density of states of the energy splitting of these 2LS.

The dynamic properties of glasses at low temperatures are governed by the strong coupling between the 2LS and elastic strains /3/. Measurements of the phonon mean free path by ultrasonic techniques have demonstrated that the resonant absorption and emission of phonons is the dominant interaction. This process not only explains the low thermal conductivity but also the temperature variation of the sound velocity and above all the power dependence of the ultrasonic absorption /2, 4/ : Below a critical intensity I_c the population of the two states is not influenced and therefore the acoustic absorption is found to be power-independent. With increasing intensity, however, both levels become more and more equally populated leading to a decrease of the acoustic attenuation, i.e. saturation occurs. For example I_c=10^-7 W/cm^2 in vitreous silica at 750 MHz and T = 0.5 K.

The 2LS not only couple to external strains, but also to each other via internal strains /3-6/ : a 2LS located at site i feels the strain field of a 2LS at site j. Because of the broad energy distribution of the 2LS and their random spatial arrangement, an interaction between 2LS of equal energy splitting is very improbable. The coupling discussed here mainly occurs between 2LS of different energy. Driven by thermal agitation transitions between the two states of a 2LS take place continuously. This transitions modulate the local strain field surrounding a 2LS and lead to a temporal fluctuation of the energy splitting of neighbouring 2LS. This interaction gives rise to a considerable broadening of the resonance line observed in ultrasonic "hole burning" experiments /6/.

Here we report on the temporal aspect of the interaction, which has not been experimentally studied so far. Since the level splitting E of a 2LS is a time dependent quantity interesting effects occur when the characteristic times of the energy fluctuations become comparable with the time of observation /7/. The fluctuation of the energy of a given 2LS depends on the relaxation time of its neighbouring 2LS.

At T = 0.5 K this relaxation time is of the order of microseconds. Ultrasonic experiments probing the energy splitting on this time scale should therefore reveal time dependent effects.

In this paper we report on the first direct experimental study of this problem : Two acoustic pulses P_1 and P_2 of variable duration (between 0.3 and 1.2 us) and of different power I (I_1>>I_c>>I_2)
were generated in a sample of vitreous silica (Suprasil I). Both pulses travel together through the sample. We measured the change of the resonant attenuation of the weak test pulse $P_2$ as a function of the frequency of the strong saturating pulse $P_1$.

The time dependence of the effect was studied by varying the width of the ultrasonic pulses applied in the experiment.

Similar to experiments carried out previously, a broad minimum in the attenuation of the probing pulse occurs at frequencies close to the frequency of the saturating pulse (see Figure 1).

The width of the line is much larger than expected from the frequency uncertainty of the ultrasonic pulses or the finite lifetimes of the 2LS.

![Figure 1](image1.png)

**Figure 1**: Variation of the resonant absorption of the weak probing pulse $P_2$ as function of the saturating pulse $P_1$ for two different pulse durations $\tau_p$.

Furthermore, the linewidth depends on the duration of the applied pulse: with increasing $\tau_p$ the line becomes wider. This result is shown more quantitatively in Figure 2 where the linewidth is plotted as a function of the pulse duration $\tau_p$.

![Figure 2](image2.png)

**Figure 2**: Observed linewidth in vitreous silica at 0.42 K as a function of the pulse duration $\tau_p$. Dashed line represents the theoretical prediction of /7/.

This experimental result can be described in terms of spectral diffusion /7/. The energy radiated into the ensemble of 2LS with an energy splitting close to the energy of the ultrasonic phonons is able to diffuse to the wings of the line during the probing time $\tau_p$. The maximum linewidth is given by the energy of interaction between the 2LS:

$$\Delta E = \hbar \Delta \nu_m = p^2/\epsilon_{ij}^2$$

where $p$ is the average elastic dipole moment associated with the 2LS and $\epsilon_{ij}$ is the distance between the 2LS located at site $i$ and $j$, respectively. A detailed analysis of the time evolution of the line has been carried out by Black and Halperin /7/ and leads to the expression:

$$\Delta \nu(t) = \Delta \nu_m f(t)$$

The function $f(t)$ describes, how the hole burnt into the population of the 2LS develops with time. It depends mainly on the distribution of the barrier heights of the double well potentials mentioned above. In figure 2 the dashed line represents a numerical calculation /7/ which is in good quantitative agreement with our results. A distribution in which the higher barriers are more pronounced would even lead to better agreement.

In summary we have demonstrated for the first time that the linewidth observed in acoustic "hole burning" experiments is time dependent in glasses. The experimental results give new information on the dynamic behaviour of the low-energy excitations present in glasses and are in good agreement with theoretical calculations.

### References


