SATURATION OF THE DIELECTRIC ABSORPTION OF
VITREOUS SILICA AT LOW TEMPERATURES

M. Von SCHICKFUS and S. HUNKLINGER [1]

*Max-Planck-Institut für Festkörperforschung and Centre de la Recherche sur les Très Basses Températures,
C.N.R.S. 166 X, F 38042 Grenoble-Cedex, France*

The dielectric absorption of vitreous silica containing 1200 ppm of OH⁻-impurities has been measured at 10 GHz
in the temperature range between 0.4 K and 8 K. A strong intensity dependence of the absorption coefficient has
been observed at temperatures below 3 K.

At low temperatures glasses exhibit anomalies which
can be attributed to low energy excitations (see for
example [1]). For example, a heat capacity in excess
of the phonon contribution is observed down to the
lowest temperatures [2,3]. The strong interaction be-
tween these low-energy excitations and phonons leads
to a very small thermal conductivity and to unusual
effects in the propagation of elastic waves. Especially
the observation of a saturation of the ultrasonic
absorption which occurs already at moderate intensi-
ties [4], shows that these excitations can be considered
as highly anharmonic oscillators – or, more conveniently
as two-level systems (2LS). In recent experiments an
anomalous variation of the dielectric constant with
temperature has been observed, and has been inter-
preted as a coupling between these 2LS and electric
fields via polar impurities like OH⁻-ions [5]. In this
paper we show for the first time that this interaction
dominates the low-temperature dielectric absorption
of glasses in the microwave range and that it leads to
a saturation effect. It should be mentioned that the
OH⁻-ions seem to form also additional low energy
states [3,6] which are similar to those caused by
paraelectric impurities in crystals [7,8]. But the
presence of these states cannot explain complementary
experiments [9]. It seems that their coupling to elec-
trical and acoustic waves is relatively weak [10],
leading only to a negligible contribution to the electri-
cal and acoustic properties of glasses at low tempera-
tures.

[1] Present address: Max-Planck-Institut für Festkörperforschung,
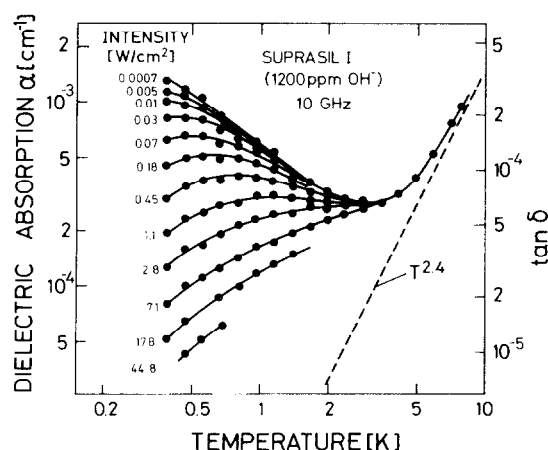D-7 Stuttgart, W.-Germany.

Fig. 1. Temperature- and intensity dependence of the dielec-
tric absorption of vitreous silica at 10 GHz. The dashed line
indicates the contribution of the relaxation process.

We have investigated the dielectric absorption of
vitreous silica Suprasil I containing 1200 ppm of OH⁻-
ions. Between 0.4 K and 8 K and at 10 GHz the absorp-
tion has been measured by determining the quality fac-
tor of a superconducting cavity resonator. In fig. 1 the
dielectric absorption is plotted against temperature
for different microwave intensities J. The absorption
is clearly seen to depend on intensity. At relatively
high temperatures and/or relatively high power levels
the absorption decreases rapidly on cooling. If the
electromagnetic intensity is small, however, the loss
passes through a minimum and increases again.

In fig. 2 the dielectric absorption at a fixed tempe-
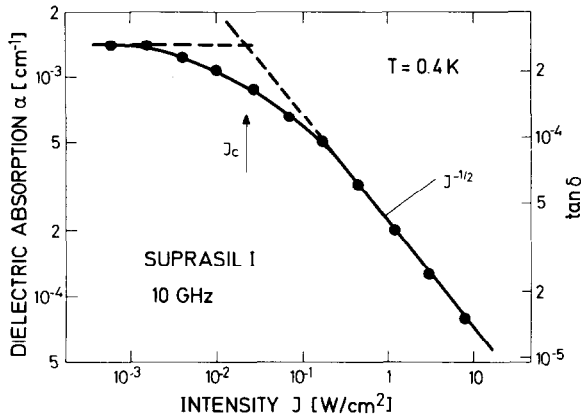rature of 0.4 K is plotted as a function of microwave

Fig. 2. Intensity dependence of the dielectric absorption at a fixed temperature of 0.4 K. Above $J = 0.1$ W/cm^2 the absorption varies like $J^{-1/2}$.

Fig. 3. Critical intensity as a function of T. Between 0.4 K and 1 K it increases almost proportional to T^4.

intensity. At higher intensities the power dependence of the absorption sets in and leads to a decrease like $J^{-1/2}$.

These experimental results can be described by the interaction between the electric field and 2LS. Without correction for the local field the Hamiltonian H of an individual 2LS in the perturbing electric field is given by

$$H = \frac{1}{2} \begin{pmatrix} E & 0 \\ 0 & -E \end{pmatrix} + \frac{1}{2} \begin{pmatrix} \mu & 2\mu' \\ 2\mu' & -\mu \end{pmatrix} F , \qquad (1)$$

where E is the energy splitting of the 2LS without perturbation, F is the external electric field and the matrix elements μ and μ' are dipole moments. We call them "permanent" and "induced" dipoles, respectively. This Hamiltonian is completely analogous to that describing the perturbation of the 2LS by a sound wave [1]. Therefore the acoustic case can be "translated" into the electric case by replacing the acoustic by the corresponding electromagnetic quantities.

With these substitutions the electromagnetic absorption coefficient α is obtained:

$$\alpha = \frac{4\pi^2 \omega n_e \mu'^2}{3 c_0 \sqrt{\epsilon}} (1 + J/J_c)^{-1/2} \tanh\left(\frac{\hbar\omega}{2kT}\right) , \qquad (2)$$

where $\omega/2\pi$ is the measuring frequency, c_0 the velocity of light in vacuum and ϵ the dielectric constant of the sample. n_e represents the density of states of those 2LS that couple to the electric field and which may be considered as energy independent [5]. The second
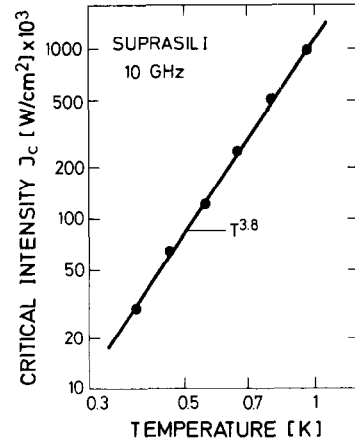
factor in eq. (2) describes the intensity dependence of the absorption and becomes important above a critical intensity J_c. The temperature dependence of the absorption is given by the last factor; it simply reflects the population difference of the two states of the 2LS caused by thermal excitation. Below 1.5 K, the measured absorption in the unsaturated regime ($J < 10^{-3}$ W/cm^2) has been fitted to this expression with the coupling constant $n_e\mu'^2$ as an adjustable parameter. The resulting $n_e\mu'^2 = 1.46 \times 10^{-4}$ is in good agreement with the value found from the temperature dependence of the dielectric constant [5].

According to eq. (2) the absorption depends on intensity above J_c and is expected to be proportional to $J^{-1/2}$ for $J \gg J_c$. As illustrated by fig. 2 such a dependence is observed for intensities well above 0.1 W/cm^2. Here the critical intensity is defined by

$$J_c = \hbar^2 c_0 \sqrt{\epsilon}/8\pi\mu'^2 T_1 T_2 , \qquad (3)$$

where T_1 and T_2 represent the lifetime of the 2LS in the excited state and the broadening of the levels due to their mutual interaction. For a quantitative analysis the magnitude of T_1 is not known well enough [1,5,11], but a value between 1 μsec and 10 nsec at 0.4 K and 10 GHz seems to be reasonable. The value of $T_2 = 3$ nsec has been measured for vitreous silica at 1 GHz and 0.4 K [12]. Under the assumption that no strong energy dependence of the mutual interaction exists we can use this value. Since $J_c = 25$ mW/cm^2 (indicated by an arrow in fig. 2), we deduce a dipole moment μ'

between 2 and 20 debye from eq. (3).

In fig. 3 the critical intensity J_c is plotted as a function of temperature. J_c and consequently also $(T_1 T_2)^{-1}$ vary almost like T^4. Since T_1^{-1} is known to be proportional to T [13, 14, 15], T_2^{-1} seems to be proportional to T^{-3}. This result is in contrast to estimates from acoustic measurements [16, 17, 18], where T_2^{-1} has been found to depend linearly or quadratically on temperature. This discrepancy and the rather low saturation intensity J_c resulting in a comparatively high dipole moment μ' could indicate that a phonon bottleneck [19] occurred in our experiment. In this case phonons emitted by a decaying 2LS could reexcite other resonant 2LS before they are thermalized by other mechanisms. This process would be in competition with the excitation by the electric field and thus account for the discrepancies mentioned above.

The interaction between the electric field and the 2LS through the "permanent" dipole moment μ results in a change of their energy splittings E. Therefore thermal equilibrium of the 2LS is disturbed by the electromagnetic wave and is restored within the relaxation time T_1 by the emission or absorption of thermal phonons. Because of the distribution of relaxation times of 2LS of different energy splitting we have to carry out an integration over all energies. If T_1 is only due to the direct process, i.e. the emission or absorption of one phonon, the condition $\omega T_1 \gg 1$ should hold in our temperature and frequency range, resulting in a T^3 law for this contribution to the absorption [1]. As can be seen in fig. 1, the contribution of the relaxation process dominates above 4K. By subtracting the resonant part of the absorption extrapolated from lower temperatures, the relaxation part is obtained. We find a temperature dependence $\alpha \propto T^{2.4}$. This result indicates that even at temperatures up to 10 K the direct process is still dominant, since all processes of higher order (Raman for example) would lead to a stronger temperature dependence of the relaxation absorption. The reason for the fact that the exponent is even smaller than 3 is, however, not yet fully understood.

In conclusion, we report here the first direct observation of an intensity dependence of the dielectric absorption in a glass. The results fit well into the general picture obtained from measurements of the dielectric constant [5] and of the ultrasonic absorption in the presence of microwave fields at low temperatures [9].

The unusually high dipole moment μ' and the strong temperature dependence of the critical intensity demonstrate, however, that the origin and the dynamics of the interaction between the 2LS are not yet understood. Apart from the importance of absorption measurements for the application of amorphous materials as low-temperature dielectrics, experiments of this type give new insight into the dynamical behaviour of low-energy excitations that determine the dielectric properties of glasses at low temperatures.

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