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EFFECT OF ELECTRON IRRADIATION ON THE HEAT CAPACITY OF VITREOUS SILICA

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Résumé.- La chaleur spécifique de la silice vitreuse (Suprasil W) à basse température (50 mK ≤ T ≤ 2 K) reste pratiquement inchangée quand l'échantillon est irradié aux électrons (3 MeV). La densité des excitations de basse énergie intrinsèque à la structure amorphe apparaît donc ne pas être affectée par l'existence de liaisons non saturées qui sont introduites dans les échantillons pendant l'irradiation.

Abstract.—The specific heat of vitreous silica (Suprasil W) at low temperatures (50 mK ≤ T ≤ 2 K) remains virtually unchanged when the sample is irradiated with 3 MeV-electrons. Hence the density of low energy excitations inherent in the amorphous structure appears not to be affected by the existence of unsaturated bonds which are introduced in the samples during irradiation.

Many of the low temperature properties of amorphous solids can be accounted for by assuming the existence of some sort of low energy excitations. These excitations are successfully described as systems in which a tunnelling process between two nearly degenerate energy levels separated by an energy E can occur /1,2/. For example the roughly linear contribution to the specific heat C observed in all glasses at low temperatures /3,4/ — in addition to the usual \( T^3 \) term — is interpreted as resulting from a roughly constant density of states \( n(E) \) of these two level systems. However, the microscopic nature of the tunnelling process, i.e. the tunnelling "entity", giving rise to two level systems is not known. These excitations are thought to be inherent in the amorphous structure. Hence, upon changing the structure of an amorphous solid in a more or less controlled way, e.g. by doping with impurities or irradiating the sample, and subsequent measurement of the specific heat and other physical properties, one might be able to draw some conclusion as to the nature of the two level systems. Up to now, the introduction of OH impurities into glass has yielded interesting effects concerning the interplay between extrinsic and intrinsic two level systems /5/, but as yet has not been able to reveal the nature of the intrinsic excitations. Consequently, it was felt worthwhile to introduce defects by electron irradiation and see if the low temperature specific heat, i.e. the density of low energy excitations was changed.

Two samples of vitreous silica (Suprasil W /6/) were irradiated by fast electrons (3 MeV) from a Van de Graaff generator. Having cylindrical shape (Ø 15 mm, h = 50 mm) they rotated slowly about their axis to ensure uniform irradiation as checked by the quite homogenous coloring of pyrex glass. One of the Suprasil W samples received a total dose of \( 8 \times 10^{17} \) e\(^{-}\), the other \( 9 \times 10^{18} \) e\(^{-}\). Neither of the samples showed coloration after the irradiation.

The specific heat \( C_p \) of the two irradiated samples as well as that of an unirradiated specimen of the same charge was measured as described in /7/.

Figure 1 shows the \( C_p \) vs. T curves for unirradiated and electron irradiated Suprasil W glass in a log-log plot. All curves show the typical behavior of glasses with \( C_p \sim T^{1.5} \) below 0.5 K as observed previously /4/ for the same type of vitreous silica. The remarkable result is that (within the error margin of ± 5 %) no difference in \( C_p \) is observed between the unirradiated and irradiated samples between 50 mK and 0.5 K, i.e. in the temperature region when the two level systems dominate in the specific heat. At higher temperatures (0.5 K < T < 2 K), \( C_p \) is systematically suppressed by up to 20 % after irradiation. However, as already the behavior of the unirradiated sample in this temperature region is complex and not fully understood, we will not discuss this effect further.

From the identity of the specific heat curves of unirradiated and irradiated samples at low temperatures we must conclude that the density of states of the low energy excitations is not changed by the structural defects which are introduced by electron bombardment. Very recently, a decrease of...
\[ C_p(\text{erg/kg}) \]

**Fig. 1**: Specific heat of unirradiated and electron irradiated vitreous silica with total electron dose of $8 \times 10^{18}$ (open circles) and $9 \times 10^{18}$ (open triangles).

$C_p$ of 40 \% was observed in vitreous silica containing about 1000 ppm OH which had been exposed to fast neutrons to a dose of up to $5 \times 10^{19}$ cm$^{-2}$ /8/.

With the aid of thermal conductivity and ultrasound measurements, the change of $C_p$ could be attributed to a decrease in the number of two level systems /8/.

Even if this change in $C_p$ is entirely due to a change of intrinsic (and not OH) two level systems, it appears to be surprisingly small. We are left with the conclusion that structural change introduced by electron or neutron irradiation, has little—if any—effect on the low energy excitations in glasses.

In order to get more information on the structural changes introduced by electron irradiation, we performed electron paramagnetic resonance measurements on our samples to obtain the order of magnitude of free spins.

No EPR signal could be detected for the unirradiated glass (corresponding to a free spin density $< 10^{14}$ cm$^{-3}$). However, for the irradiated samples we observed a spin density of order $10^{17}$ cm$^{-3}$, i.e. of the same order as the number of intrinsic two level systems. Hence, from this experiment in conjunction with the above specific heat results of the same samples we conclude that unsaturated bonds, i.e. free spins do not have a significant effect on the low energy excitations in glass. This seems to rule out models invoking motion of single electrons or broken bonds.

In conclusion the density of low energy excitations is not changed by electron irradiation up to a total dose of $\sim 10^{19}$. Hence, these excitations are unaffected by presence or absence of unsaturated bonds. Perhaps with more experiments of the kind reported here one gradually can pin down the mechanism which gives rise to low energy excitations in amorphous solids.

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