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Comments and Addenda

The section Comments and Addenda is for short communications which are not appropriate for regular articles. It includes only the following types of communications: (1) Comments on papers previously published in **The Physical Review** *or* **Physical Review Letters**. *(2) Addenda to papers previously published in* **The Physical Review** *or* **Physical Review Letters**, *in which the additional information can be presented without the need for writing a complete article. Manuscripts intended for this section must be accompanied by a brief abstract for information-retrieval purposes. Accepted manuscripts follow the same publication schedule as articles in this journal, and page proofs are sent to authors.*

Effect of neutron irradiation on the low-temperature dielectric susceptibility of vitreous silica

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Whereas neutron irradiation changes the apparent density of low-energy localized excitations in vitreous silica as monitored by measurements at temperatures below 1 K of thermal conductivity, specific heat, and temperature dependence of the ultrasonic velocity, no change is detected in the density of localized excitations which contribute to the dielectric susceptibility.

In a previous publication[1] we discussed measurements of the thermal conductivities κ, the temperature dependencies of the ultrasonic velocities $\Delta v/v$, and the excess specific heats C_{ex} of vitreous silica and of neutron-irradiated vitreous silica. These measurements were made in a study of the localized low-energy excitations which occur in amorphous materials. The changes induced in C_{ex}, κ, and $\Delta v/v$ below 1 K by neutron irradiation could be explained by a 35% reduction in the density of localized excitations. The present paper discusses the effect of neutron irradiation on the temperature dependence of the dielectric constant ϵ of vitreous silica. We find, in brief, that neutron irradiation does *not* change the density of of those localized excitations which contribute to ϵ.

The temperature-dependent dielectric constants of three samples were measured; all three were originally cut from the same piece of Spectrosil-*B* vitreous silica[2] used in Ref. 1. One sample was measured in the unirradiated condition. The second sample had been neutron irradiated for "30 days"[3] at Argonne National Laboratory and is identical to the samples used in Ref. 1, having an apparent 35% reduction in the density of localized excitations. The third sample had been neutron irradiated at the Research Reactor of the University of Missouri, Columbia. This third sample had an apparent reduction of 30% in the density of localized excitations as deduced from measurements of κ. Due to indications that the dielectric properties of vitreous silica are closely related to the concentration of OH^- impurities, the characteristic OH^- absorption band at 2.73 μm was monitored. All three samples were found to have the same OH^- content, which was roughly 1000 ppm by weight.[2]

The technique used in the measurements of ϵ has been described in Ref. 4. The results of the measurements are shown in Fig. 1, where the di-

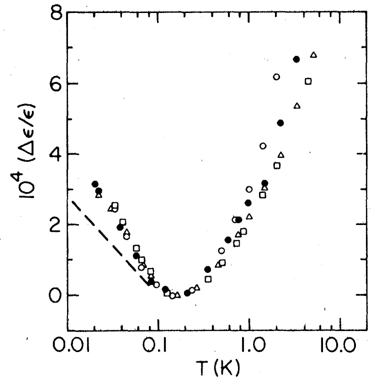

FIG. 1. Variation with temperature of the dielectric constant of vitreous silica measured at 10^4Hz. △, neutron irradiated (Missouri); □, neutron irradiated (Argonne); ○, ●, unirradiated, two runs. Roughly half the data have been omitted for clarity. The dashed line is discussed in the text.

electric constant is plotted as $\Delta\epsilon/\epsilon = [\epsilon(T) - \epsilon_{\min}]/\epsilon_{\min}$, with $\epsilon_{\min}$ equal to the minimum value of ϵ. The present data are in agreement with earlier data[5,6] for unirradiated vitreous silica having a similar OH$^-$ concentration.

No difference between the irradiated and unirradiated samples can be detected in Fig. 1 for $T \leq 0.4$ K. Since the temperature dependence of ϵ is proportional to the density of localized excitations active in dielectric behavior (see below), the data indicate that the density of electrically active excitations is not changed by neutron irradiation. The effect of a reduction in the density of excitations comparable to that deduced from C_{ex}, κ, and $\Delta v/v$ is indicated by the dashed line in Fig. 1. At temperatures above ≈0.4 K the irradiated material appears to exhibit a temperature dependence ≈25% smaller than the unirradiated material. However, the data were less reproducible above 1 K, as indicated by the two runs on unirradiated samples. For this reason we will focus on data obtained below 0.4 K. It is, in fact, the lowest-temperature data which most accurately reflect the density of localized excitations having lowest energies.

Previous work on the low-temperature dielectric constant of vitreous silica containing OH$^-$ impurities has indicated that the electrical properties of the material are determined by the OH$^-$ impurities, and that the number of electrically active localized excitations scales linearly with the OH$^-$ impurity concentration.[6] Measurements of the low-temperature thermal properties of silicas having different OH$^-$ contents suggest that OH$^-$ may affect the density of localized excitations active in the excess specific heat, while leaving the density active in the thermal conductivity essentially unaltered.[7] However, it should be emphasized that attempts, such as described above, to attribute differences in the thermal properties of samples to different OH$^-$ concentrations may not be valid—the different OH$^-$ levels also reflect different sample histories.

The exact relationship between localized excitations related to OH$^-$ impurities and localized excitations intrinsic to pure silica is unknown. That is, it is not known whether the OH$^-$ produces excitations independent of the "intrinsic" ones, or whether the OH$^-$ decorates the sites of the intrinsic excitations, thus electrically activating them. Simultaneous electromagnetic and acoustic measurements have indicated that the OH$^-$-related and intrinsic (strong phonon scattering) excitations are strongly coupled.[8,9] Additionally, the observation that the temperature dependence of the dielectric constant scales linearly with the OH$^-$ concentration suggests that if the OH$^-$ ions electrically activate existing excitations, then there is, at most, one OH$^-$ ion per localized excitation.[6]

Our earlier data[1] on C_{ex}, κ, and $\Delta v/v$ could be accounted for quantitatively by a model[10,11] which ascribes the localized excitations to the tunneling of an atom or larger entity between two potential-energy minima having similar ground-state energies. A distribution of two-level systems results from the tunneling. This tunneling-states model may also be applied to the present data to provide an estimate of the density of electrically active excitations. The contribution of resonant absorption by two-level systems to the variation in the dielectric constant is given by[6]

$$(\Delta\epsilon/\epsilon)(T) = (8\pi/\epsilon)P_e\mu'^2 \ln(T/T_0)\,, \tag{1}$$

where P_e is the number of electrically active two-level systems per unit energy interval, per unit volume. Here μ' is the electric-dipole matrix element coupling the ground and excited states of the two-level system. Ths value of ϵ for the silica[12] is about 3.8. From Eq. (1) and the slope of the low-temperature $\Delta\epsilon/\epsilon$ curve of Fig. 1 we find that

$$\mu'^2 P_e = 3.3\times10^{-5}\,. \tag{2}$$

The value of μ' has been determined from electric echo experiments.[13,14] Although the experimental values differ somewhat, a conservatively small value[15] is believed to be about 1 D. Using 1 D in Eq. (2), we find $P_e = 3.3\times10^{31}$ erg^{-1} cm^{-3}. It was found in Ref. 1 that the density of states P active in the thermal and acoustic measurements is about 7×10^{32} erg^{-1} cm^{-3} for the irradiated material (smallest P). Hence, at most, the electrically active states comprise about 5% of the states active in the thermal and acoustic properties.

This small fraction of electrically active states could explain the fact that large electric fields have no effect on the thermal conductivity of vitreous silica.[16,17] If the same total dipole moment were ascribed to *all* the states present in the glass, the average dipole moment per excitation would be 0.2 D, a value consistent with the result of Stephens.[15] Also, since the fraction of electrically active states is so small, any difference in their contributions to κ, C_{ex}, or $\Delta v/v$ could not have been detected in Ref. 1.

The 1000-ppm OH$^-$ content of the vitreous silica used in the present measurements corresponds to an OH$^-$ concentration of about 8×10^{19} cm^{-3}. If it is assumed that the two-level systems related to the OH$^-$ have splitting energies ranging from zero to 30 K, this OH$^-$ concentration would yield an OH$^-$-related density of states of 2×10^{34} erg^{-1} cm^{-3}, more than 600 times the observed electrically active density of states. Hence the OH$^-$ ions appear to be very inefficient at providing electrically active two-level systems.

In summary we find that neutron irradiation of vitreous silica does not alter the density of localized excitations which contribute to the dielectric susceptibility; this is in contrast to the irradiation-induced changes in the density of excitations which dominate the low-temperature specific heat, thermal conductivity, and ultrasonic dispersion. In addition, the electrically active excitations represent only a small fraction of the total density of localized excitations.

ACKNOWLEDGMENT

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