Very-low-frequency light scattering in silica at low temperatures

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We report a study of the light scattered at low-frequency shifts (0.03–20 cm^{-1}) in a silica-core glass fiber at temperatures as low as 1.46 K. Although our data agree in regions of spectral overlap with those reported previously, they provide a more detailed examination extending down to ~0.03 cm^{-1} (including the Brillouin spectrum). Contrary to extrapolations by earlier workers based on higher-frequency data, we find no evidence of a central component due to scattering from two-level tunneling systems.

The subject of low-frequency light scattering in glasses has been an intriguing one for some time, largely because of the apparent possibility of studying directly the two-level tunneling systems (TLTS) responsible for the anomalous low-temperature thermal properties of glasses.[1] This scattering can be either direct or indirect via relaxation. Although the direct scattering intensity due to TLTS has been predicted[2] to *increase* at low temperatures, the low-frequency scattering observed in various glasses[3–5] exhibits a *decreased* intensity as the temperature is reduced. In particular, a low-frequency "excess" scattering intensity in fused silica has been attributed[3,6] to relaxation process involving TLTS, but the region of temperature where the predicted increase in the *direct* scattered intensity should occur has only recently been explored.[6] The region of interest is this regard is $kT < h\nu$.

The recent attempt to observe the increased direct scattering at low temperatures employed a silica-core fiber[6] to provide long path length. The fiber, about 330 m in length, was supported in a strain-free fashion by coiling in a thin-wall metal trough suspended in a liquid-He bath by fine wires. A multipass Fabry-Perot interferometer in tandem with a grating spectrometer was used to measure isolated points in the spectrum down to 2.0 cm^{-1}. In the present work, we have used the same fiber and Dewar arrangement as in Ref. 6, but with the low-frequency scattering technique based on the use of an iodine reabsorption cell,[7] which permits direct observation of spectra down to $\Delta\nu \cong 0.03$ cm^{-1}. Although our data do not conflict with those previously reported, the new information on the low-frequency portion of the spectrum ($\nu < 1$ cm^{-1}) leads us to conclude that there is no evidence for a central component due to scattering from two-level tunneling systems.

Stolen and Bösch obtained data down to a frequency shift of $\nu \sim 2$ cm^{-1}. The results agree in essence with those of Winterling[3] where they overlap at higher temperatures. At low temperatures (~4.2 K) the data suggest a narrowing of the intensity seen by Winterling, but at the lowest temperature (1.5 K) no evidence was found for the markedly increased intensity predicted theoretically.[2] The present investigation was undertaken, then, for several reasons: (1) To investigate the very low-frequency region of the spectrum (down to $\nu \sim 0.03$ cm^{-1}) for evidence of the narrow central peak suggested previously;[2,3,6] (2) to evaluate the contribution of Brillouin scattering to the spectrum in the 3–5-cm^{-1} region; and (3) to repeat the measurements of Winterling[3] by a different technique.

The incident beam is ~10–50 mW of 5145-Å radiation from a single-mode Ar^+ laser, stabilized at the I_2 absorption line.[7] Two experimental geometries are employed, shown schematically in Fig. 1. Actually, both Fabry-Perot and grating spectra were taken in both forward and backward scattering, but, as shown in the figure, we shall emphasize here the Fabry-Perot results for backscattering and the grating spectra for forward scattering. For the respective cases the spectra are analyzed by a Spex 1401 double-grating monochromator (for $\nu > 2$ cm^{-1}, $\Gamma_i \sim 1$ cm^{-1}) or by a tandem pressure scanned Fabry-Perot interferometer[7] (for $0.03 < \nu < 2$ cm^{-1}, $\Gamma_i \sim 0.06$ cm^{-1}). As in Ref. 6, the fiber is immersed in a pumped liquid-He bath for the low-temperature studies, and the bath pressure is monitored to give actual temperatures. The temperature is controlled by adjusting the pumping speed. The fiber is a step index design, with a pure silica core (diameter 5.85 μm) and a borosilicate cladding ($\Delta n = 0.0033$), and it sustains three or four mode groups at our wavelength of $\lambda = 5145$ Å. The loss in the green is about 10 dB/km. Mode strippers are employed at both ends of the fiber to strip all intensity out of the cladding.

From the observed throughput at room temperature we estimate an insertion loss of some 80–85%,

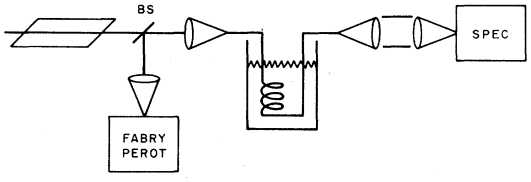

FIG. 1. Schematic of the geometries used for the present investigation. The beamsplitter (BS) separates the backscattered light from the incident beam. The forward scattered light is analyzed by a double grating spectrometer (SPEC) and the laser (at left) is a single-mode Ar^+ laser, operated at 5145 Å.

so that, after the beam splitter, the actual excitation level is in the range 1–5 mW. The increase in attenuation of some 6–8 dB observed on cooling is also larger than observed previously,[6] which may be due to preferential excitation of higher-order modes. We have also considered the possibility that stimulated Brillouin scattering may distort our results.[8] From the data of Vacher[9] and the equations of Ref. 8 we find that that threshold falls from ~50 mW near 100 K to ~5 mW near 4.2 K. (Note that the use of a single-frequency laser decreases this threshold substantially from the values for a multimode laser.[8]) Thus, at 4.2 K, if our estimate of insertion loss is correct, we are in fact quite close to threshold. However, we observe no evidence for any stimulated processes when the beam power is varied over the range indicated above, especially at 4.2 K, where the largest amount of data was taken.

Most of the data were taken at 4.2 and 1.46 K. At higher temperatures (100 K, 200 K, and room temperature) the results agree qualitatively with the detailed observations of Winterling,[3] and we shall not discuss them further here. We reduced the spectra in the same manner as did the previous workers[3,6] for the purposes of this comparison. At low temperature, the room-temperature portion of the fiber (~0.5 m) gave a noticeable contribution, but by reducing the length of that section to a minimum it was possible to make this contribution small. Moreover, in the backscattered geometry it was possible to reject completely the polarized component (the LA mode) of the scattered light from the room-temperature portion of the fiber.

This rejection was particularly important in the case of the Fabry-Perot spectra. While it did not assist in rejection of the (unpolarized) low-frequency density-of-states scattering, it did greatly improve the rejection of the secondary *forward* Brillouin scattering observed in the *backscattering* geometry, excited by elastically backscattered laser light. A pair of representative 4.2-K spectra are shown in Fig. 2. Since the Fabry-Perot spectra reported here were dominated by the spectrally sharp LA modes, no attempt was made to use the normalization procedure previously reported,[7] and the Fabry-Perot spectra shown are *uncorrected*.

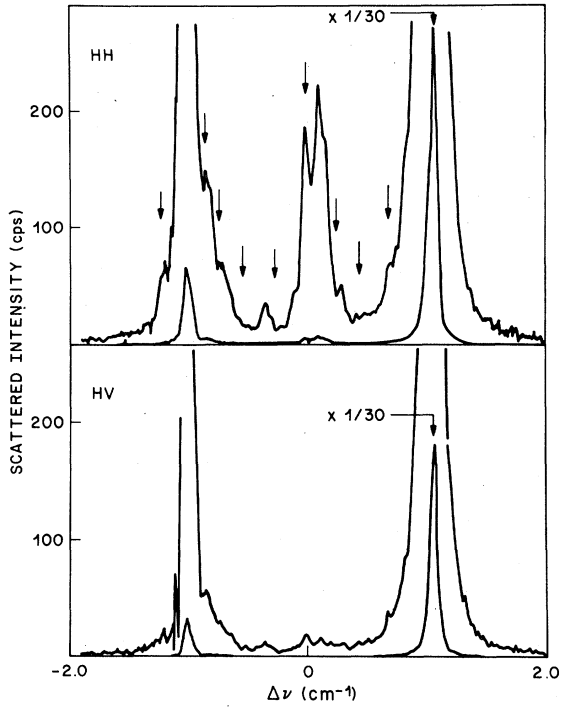

FIG. 2. Backscattered Fabry-Perot spectra obtained in polarized (*HH*) and depolarized (*HV*) geometries at 4.2 K. The spectra, shown uncorrected here, exhibit the characteristic structure due to the subsidiary I_2 absorptions indicated by the arrows, but the dark count is subtracted. The feature near $\nu \sim 0$ in the *HH* spectrum is due to forward Brillouin scattering excited by elastically backscattered laser light.

The forward-scattered component ($\nu \lesssim 1$ GHz) evident in the *HH* polarization is largely removed in *HV*. We can naturally assume that the backscattered LA contribution from the room-temperature portion of the fiber is likewise rejected efficiently. Thus, the intensity near $\nu \sim 0$ in the *HV* geometry stems mainly from the cold portion of the fiber. The Brillouin peaks here are attenuated by the iodine cell at least twice as much as the scattering at ~0.03 cm^{-1}. Thus, the observed *HV* intensity at $\nu \sim 0.03$ cm^{-1} is about 0.1% of that at the backscattered LA mode ($\nu \cong 1.17$ cm^{-1}).

In considering whether this intensity could be due to the LA tail, we assume a quasiharmonic response

$$I(\nu) = \frac{2\Gamma}{(\nu^2 - \nu_0^2)^2 - 4\Gamma^2\nu^2} \quad , \tag{1}$$

where $\Gamma \cong 0.005$ cm^{-1} is estimated from direct measurements of the LA width,[9] and ν_0 is the LA frequency. The intensity ratio observed than must be corrected for our resolution [which increases $I(\nu=0)$ relative to (ν)], and we obtain the ratio of the expected intensities:

$$I(0)/I(\nu_0) \cong 2\Gamma\Gamma_i/\nu_0^2 \cong 7 \times 10^{-4} \quad .$$

Thus, this estimate accounts for at least 70% of the observed intensity. We therefore conclude that most of the intensity observed at $\nu \to 0$ is due to LA scattering.

However, at high-frequency shifts ($\nu \gtrsim 2$ cm^{-1}), the situation is different. The ratio of the intensity at $\nu = 4$ cm^{-1} to that at $\nu \sim \nu_0$, for example, is approximately given, assuming the response (Γ), by $2\Gamma\Gamma_i\nu_0^2/\nu^4$, which gives a ratio of $\sim 5 \times 10^{-6}$. On the scale of the Fabry-Perot spectra, such an intensity is unobservable (corresponding to $\sim 10^{-2}$ cps). In fact, no clearly defined intensity was observed outside the region of the LA mode. Thus, the Fabry-Perot spectra indicate that the observed intensity at $\nu \gtrsim 2$ cm^{-1} arises from processes other than LA Brillouin scattering.

A similar analysis can be carried out for the grating spectra. In this case, it is necessary to use forward scattering (as done by Stolen and Bösch[6]) in order to avoid a large unpolarized contribution from the room-temperature portion of the fiber. For the grating spectrometer, $\Gamma_i \cong 1$ cm^{-1}, so that the expected intensity ratio at $\nu \cong 4$ cm^{-1} is $\sim 5 \times 10^{-5}$, if we assume the only contribution is from a response of the form (1). The observed intensity ratio, seen in the spectra in Fig. 3, is $\sim 10^{-2}$, more than two orders of magnitude too large to be due to the Brillouin tail. Furthermore, if we introduce a relaxing self-energy into the response (1) in the usual fashion[10] we find that in order to obtain the observed intensity at 4 cm^{-1}, we must assume a coupling strength which would renormalize the LA phonon frequency by

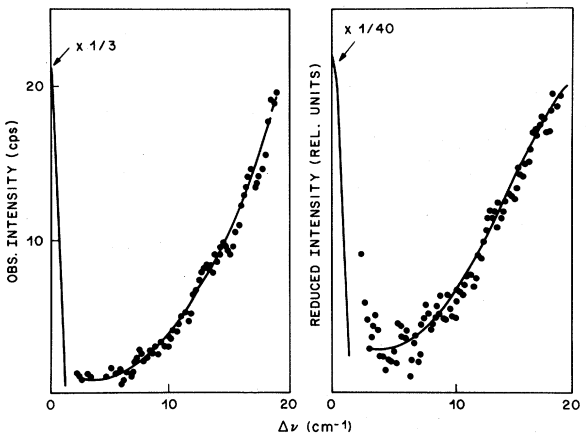

FIG. 3. Forward-scattering unpolarized grating spectra at 4.2 K, shown both as observed and as reduced by previous workers[3,6] {$I_r(\nu) = I(\nu)/[\nu n(\nu,T)]$, where n is the Bose-Einstein factor}. The integration time is ~200 sec per point. The dark count is subtracted in both cases, and the structure imposed by the I_2 absorptions is removed. The low-frequency peak is due to Brillouin scattering. The solid curves are guides to the eye.

some 10% and require an intensity near $\nu = 0$ far greater than that observed in the Fabry-Perot spectra. Hence, we concur with the argument advanced by Winterling[3] that the excess scattering in the 4–10-cm^{-1} region is not due to a Brillouin tail. Moreover, we have extended the argument to the case of an acoustic response containing a relaxing self-energy. Note that this argument does not depend upon polarization selection rules.

Let us now consider whether the excess intensity observed at low frequency exhibits the characteristic of a narrowing central component as the temperature is lowered. We first note that our grating spectra at 4.2 K (Fig. 3) do not show any evidence of a peak in the real scattering intensity as $\nu \to 0$. This finding appears to be within the error bars of the previously reported spectra.[6] Second, we can compare the intensity at $\nu \cong 0.03$ cm^{-1} in the Fabry-Perot spectra with that observed in the grating spectra. At 4.2 K, the intensity ratio $I(0.03\text{ cm}^{-1})/I(\nu_0)$ is $\sim 10^{-3}$ (Fig. 2). Of this, 7×10^{-4} is accounted for above by the quasiharmonic response of the LA mode. Correcting the remainder for the difference in resolution (the values of Γ_i) we obtain a ratio of $\sim 0.6 \times 10^{-2}$ for the grating case. Since that is close to the value of the intensity observed at 4 cm^{-1} (a ratio of 10^{-2}, as described above and shown in Fig. 3), we conclude that the observed intensity in this region in fact does *not* represent the tail of any much narrower feature, but rather is a comparatively flat background over the range 0–10 cm^{-1} at 4.2 K.

Similar arguments can be advanced relative to the 1.46-K spectra. The spectra are noisier, due to the reduced signal at this lower temperature, and therefore the conclusions cannot be drawn as quantitatively. However, within the experimental accuracy, they are the same. In particular, no increase in intensity is observed at very low-frequency shift (~ 0.03 cm^{-1} $\ll kT$), nor at large shifts (~ 3 cm^{-1} $\gtrsim kT$), in disagreement with the expectations based on the theory of Jäckle[2] if the room-temperature scattering is due to relaxation associated with two-level tunneling systems. Thus, since we clearly have data in the important region $kT \lesssim h\nu$, where such scattering is observed at higher temperatures, we conclude that either (1) the scattering observed at high temperature is not due to two-level tunneling systems or (2) the approximations employed by Jäckle are inappropriate for the present case. In either case, the theory[2] is clearly inapplicable to the description of the excess scattering intensity observed.

The scattering described here is also of a very different nature from that reported in the heavy metal oxide glasses.[5] In that case, narrow central components (3–7 GHz) observed near room temperature were observed to disappear gradually as the temperature was lowered, with little change in spectral shape. In one series of such glasses, the intensity was pro-

portional to Tb^{3+} content. No satisfactory explanation has yet been advanced for these data, but there appears to be no reason to relate those observations to the silica data presented here.

In conclusion, by obtaining grating and Fabry-Perot spectra of silica at low temperature and high resolution, with the elastic scattering attenuated by an iodine reabsorption cell, we find spectra which support Winterling's essential observation of a nonzero intensity as $\nu \rightarrow 0$. We also have extended his argument that this intensity is not the result of a wing on the LA phonon line to include the case of a relaxing self-energy contribution to the acoustic-mode response. In addition, we have checked directly and in considerable detail for the narrow component at low temperature suggested (by extrapolation) by Winterling and predicted theoretically.[2] It is not observed. We find instead that the spectrum at 4.2 K in the 0–10-cm^{-1} region consists of the quasiharmonic LA modes superimposed on a relatively flat background which is weaker by a factor of about 10^5, in terms of real spectral power density, but with an integrated intensity of some 1% of that of the LA modes. Since no anomaly is observed at low temperature ($kT \lesssim h\nu$) and at low frequencies ($\nu < 1$ cm^{-1}) we conclude, however, in contrast to earlier reports,[3,6,2] that there is no evidence to relate the observed scattered intensity to the low-temperature tunneling properties of the glassy state.

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