Infrared and Raman Study of Two-Level Systems in Fiber Optic Quality a-SiO_2 and a-SiO_2:GeO_2

B. E. Hubbard, N. I. Agladze, J. J. Tu and A. J. Sievers

Laboratory of Atomic and Solid State Physics and Cornell Center for Materials Research
Cornell University, Ithaca, NY 14853-2501

Nearly twenty years ago precise and systematic studies of the low-frequency and low-temperature Raman scattering in very pure silica fibers [1] showed that this spectroscopic probe could not identify two-level systems (TLS) in vitreous silica. These experiments failed to show any evidence of the predicted temperature dependent signature associated with the resonant TLS scattering contribution, even though this TLS feature has been observed in a variety of glasses using many different experimental probes. In our work the absence of Raman activity is reexamined with the systematic measurement of the low frequency, low temperature Raman spectra and far-infrared absorption of optical fiber quality silica-germania glasses. If the Raman activity of the TLS in a-SiO_2 involves a hidden selection rule then the large mass defect produced by the addition of the more Raman active GeO_2 should increase the vibrational disorder. If the Raman activity is simply too weak then the addition of GeO_2 should also enhance the Raman activity of the entire vibrational spectrum. For $(SiO_2)_{0.88}(GeO_2)_{0.12}$ and $(SiO_2)_{0.63}(GeO_2)_{0.37}$ glasses the low-temperature vibrational Raman scattering (including the Boson peak region) is measured to be two and three times larger than that observed for pure SiO_2. In neither case has the temperature dependent signature associated with the resonant TLS Raman scattering been observed. When the low temperature far-infrared transmission of pure fiber quality a-SiO_2 is measured, a very weak TLS signature is observed with a maximum temperature-induced change in the absorption coefficient ~ 1.9×10^{-3} cm^{-1}. The corresponding temperature-induced change for a $(SiO_2)_{0.955}(GeO_2)_{0.045}$ glass is ~ 3.8×10^{-3} cm^{-1}. Since OH is absent from these glasses these values are sufficiently small that they may be associated with other unwanted impurities. The absence of TLS Raman activity and the very weak TLS far-IR activity observed for these ultrapure silica-germania glasses suggests that the TLS tunneling does not alter the local polarizability or the dipole moment in such perfect covalently-bonded glasses.

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1. R. H. Stolen and M. A. Bösch, Phys. Rev. Lett. **48**, 805 (1982).