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Ultra-Low Temperature Properties of Amorphous and Glassy Materials

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Abstract

During the grant period we made detailed studies of the dynamics of two level tunneling systems in glasses at very low temperature and by the application of AC and DC electric fields. Models have been developed that now account for both the formation and subsequent breaking of resonant tunneling pairs, and strongly bound pairs in a swept electric field. Perhaps most importantly, we saw a critical field in the polymeric glass Mylar, beyond which recovery following the application of a strong electric field is substantially modified from the predictions of current models. It was essential during the final grant period to see how general these new properties were by testing for them in a new and broader set of glasses. At the same time, the discovery that tunneling systems with nuclei possessing electric quadrupole moments that couple the TS behavior to magnetic fields was studied in this laboratory, using some of the probes that we alone employ. Finally, we were developing our own dielectric pulsed echo system, operating for the first time at the low energy splittings and hence temperatures at which interactions between TS are important. We combined this technique with the sudden application of both electric and strain fields to better understand the dynamics of the response of TS in glasses on a much shorter time scale than is possible with our established probes.

Project Summary

The goal of this research program was to better understand the properties of amorphous and glassy solids at low temperatures. It had long been believed that these materials all displayed a universal set of anomalous low temperature properties that are independent of how the material was created and the nature of its constituents. This set of anomalous properties was believed to be well described by a theory that postulated a broad distribution of non-interacting two level tunneling systems (TS model) within glasses. It now appears that this is not so, and many of the long-held beliefs about the nature of 'low temperature glassy behavior' are simply not true. For example, the low temperature dielectric loss is much larger and decreases much more slowly with decreasing temperature than theory has predicted. In addition, the ratio of the slopes of both the dielectric constant ($\varepsilon'(\omega, T)$) and sound velocity vs. ln(T) at low and high temperatures is observed to be near minus one in most amorphous systems, rather than the factor of minus two predicted from the theory. Many of the departures from the non-interacting theory of glasses based on the existence of a broad spectrum of two level tunneling systems in glasses now seems explicable in terms of interactions between these TS, although the strength of these interactions appears to vary widely between different amorphous solids. For example, Mylar, an amorphous polymer, exhibits behavior which appears to be much better described by the non-interacting TLS theory than any other amorphous system studied so far.

The extent to which recent studies show behavior that contradicts the predictions of the non-interacting TS model continues to grow, even thirty years after the development of this theory. For example, it had long been believed that the dielectric response of amorphous solids at low temperatures was virtually independent of the magnitude of any applied magnetic field. Measurements by G. Frossati on OH^- doped fused silica certainly were consistent with that assumption. Recent measurements of multi-component glasses, however, show a bizarre field dependence on magnetic field.

The existence of a dipolar gap in the density of states of TLS in amorphous solids, first discovered in this lab, has provided a framework for understanding much of the departure of the properties of amorphous solids at low temperatures from the standard non-interacting model. During the past period we continued to study the behavior and implications of this dipolar gap. Here, we focused on the extent of the interactions, both their dynamics in the destruction of strongly bound TS pairs by a DC electric field and the subsequent formation of the same pairs once the field is returned to zero.

Once my graduate students, completed the measurements of the magnetic field dependence of the dielectric constant in a variety of glasses, we wanted to make a systematic study of the dielectric constant in mylar and *related* polymers. We have found in the past that virtually all glassy materials except mylar show behavior that violates the two-level tunneling model of low temperature glassy behavior. Specifically, all glasses measured except mylar exhibit a ratio of dependences of e'(w) vs. log(T) above and below the minimum in dielectric constant that is nearly unity, while the theory suggests that the low temperature slope should be double the high temperature slope. In addition, we find that the dielectric constant of mylar continues to rise logarithmically down with decreasing temperature to 480 microKelvin, while all other glasses show a saturation below about 2500 microKelvin. This includes several other polymers, such as kapton and various photo-resists. We also looked at the magnetic field dependence of the dielectric constant of mylar. We had not put mylar in the sample chamber, but when we had to warm up, it was added.

The results presented in this work are consistent with previously made observations that nuclear spins greater than ½ play a crucial role in the observed magnetic field dependence.

Publications

1. B. Tigner, D.J. Salvino, S. Rogge, and D.D. Osheroff, Proc. 7th Int'l Conf. Phonon Scatt. in Cond. Matt., Cornell, ed. M. Meissner and R.O. Pohl, 285 (Springer-Verlag, Berlin, 1993)

2. Sven Rogge, D.J. Salvino, B. Tigner, and D.D. Osheroff, "Low Temperature Time and Electric Field Dependence of the Dielectric Constant in Amorphous Materials", Proc. 20th Int'l Conf. on Low Temp. Phys., Physica B, **194-196**, 407 (1994).

3. D.J. Salvino, S. Rogge, B. Tigner, and D.D. Osheroff, "The Low Temperature AC Dielectric Response of Glasses to High DC Electric Fields", Phys. Rev. Lett. **73**, 268 (1994).

4. D. D. Osheroff, Sven Rogge, and Douglas Natelson, "Anomalous Dielectric Properties of Amorphous Solids at Low Temperatures", Proceedings of the Combined Conference of the 4th International Conference on Phonon Physics and the 8th International Conference on Phonon Scattering in Condensed Matter, Physica B **219&220**, 243 (1996).

5. Sven Rogge, Douglas Natelson, and D.D. Osheroff, "Evidence for the Importance of Interactions Between Active Defects in Glasses", Phys. Rev. Lett. **76**, 3136 (1996).

6. Douglas Osheroff, Sven Rogge, and Douglas Natelson, "Interactions Between Active Defects in Glasses at Low Temperatures", Proceedings of the 21st International Conference on Low Temperature Physics, LT-21, Czech J. Phys. **46**, Suppl. S6, 3295 (1996).

7. Sven Rogge, Doug Natelson, and D.D. Osheroff, "Non-equlibrium and Hysteretic Low Temperature Dielectric Response to Strain in Glasses" JLTP, **107**, 717 (1997).

8. Sven Rogge, Douglas Natelson, and D.D. Osheroff, "Anomalous Behavior of e(w) in Glasses at Low Temperature Due to Bias Application", Proceedings of the 21st International Conference on Low Temperature Physics, LT-21, Czech J. of Phys. **46-Supp**. 2263 (1996).

9. Douglas Natelson, Sven Rogge, and D.D. Osheroff, "Dielectric Response of Two Level Systems to Strain Fields at Low Temperatures", Proceedings of the 21st International Conference on Low Temperature Physics, LT-21, Czech J. of Phys. **46-Supp**. 2265 (1996).

10. Sven Rogge, D. Natelson, B. Tigner, and D. D. Osheroff, "Non-linear Dielectric Response of Glasses at Low Temperature", Phys. Rev. B **55**, 11256 (1997).

11. Sven Rogge, D. Natelson, B. Tigner, and D. D. Osheroff, ^{3}He Immersion Cell for Ultralow Temperature Sudy of Amorphous Solids", Rev. Sci. Inst., **68**, 1831 (1997).

12. S. Rogge, D. Natelson, D.D. Osheroff, "Non equilibrium and Low Temperature Dielectric Response to Strain in Glasses", J. Low Temp. Phys. **106**, 717-25 (1997).

13. "Interactions Between Tunneling Defects in Solids, a chapter of a new book by Springer Verlag Alexander Burin, Douglas Natelson, Douglas D. Osheroff, and Yuri Kagan, entitled "Tunneling systems in solids" (Feb. '98). edit. by Pablo Esquinazi, Universitat Leipzig.

14. D. Natelson, D. Rosenberg, and D.D. Osheroff, "Evidence for Growth of Collective Excitations in Glasses at Low Temperatures", Phys. Rev. Lett. **80**, 4689, (1998).

15. Danna Rosenberg, Douglas Natelson, and D.D Osheroff, “Thermal Conductivity in Glasses Below 1 K: New Technique and Results”, J. Low Temp. Phys., **120**, 259-268 (2000).

16. D. Rosenberg, P. Nalbach and D.D. Osheroff, “Memory Effects in Amorphous Solids Below 20 mK”, Phys. Rev. Lett. **90**, 195501 (2003).

17. S. Ludwig, P. Nalbach, D. Rosenberg, and D. Osheroff, “'Dynamics of the Destruction and Rebuilding of a Dipole Gap in Glasses”, Phys. Rev. Lett. **90**, 105501 (2003).

18. S. Ludwig and D. D. Osheroff, “Field-Induced Structural Aging in Glasses at Ultralow Temperatures”, Phys. Rev. Lett. **91**, 105501 (2003).

19. P. Nalbach, D. Osheroff and S. Ludwig, ‘Non-equilibrium Dynamics of Interacting Tunneling States in Glasses’, J. Low Temp. Phys. **137** 3/4, November 2004.