Chapter 5

Conclusions and Future Work

5.1 Conclusions

We used dielectric spectroscopy to measure the frequency and temperature dependence of the low frequency dielectric function $\varepsilon(\omega)$ of thin C_{60} films. We found that oxygen rapidly diffuses into the grain boundaries of the C_{60} polycrystalline film, increasing its resistivity by about seven orders of magnitude. Water, on the other hand, does not penetrate into the bulk of C_{60} solid, and the increase in conduction resulting from its presence on the surface of a C_{60} film is fully reversible.

In order to explain the Debye-like dielectric loss peak and the accompanying increase in polarization observed at $\sim$35 Hz, a small charge transfer is inferred between the C_{60} and O_2 molecules which occupy interstitial spaces of C_{60} solid exposed to oxygen (or ambient air). Due to the large size of the C_{60} molecules, this small charge transfer creates large dipole moments, which in turn are coupled to the applied AC electric field via a *diffusion-controlled relaxation* mechanism. These dipole moments are responsible for a significant increase of permittivity ε' at low frequencies.

With increasing oxygenation, the interstitial sites become nearly fully occupied, interstitial hopping is inhibited, and the loss peaks, together with the enhanced polarizations, disappear.

We note that C_{60} can be thought of as a rather potent "charge sponge." In M_3C_{60} (M=alkali metal), this leads to fascinating superconducting compounds (Sec. 1.5); in thin-film $\{Al\text{-}C_{60}\}_n$ multilayer structures, charge transfer leads to interesting interface effects [273]; and in our study, charge transfer leads to extraordinary additional polarization.

We conclude that dielectric and, especially, microdielectric spectroscopy can be used to assess the content of oxygen or other impurities in the interstitial spaces of C_{60} solid. Both tracer and chemical diffusion coefficients can be determined. We have used this technique to ascertain the effect of various intercalants on the dielectric properties of C_{60} films.

5.2 Possible Applications

The original goal of our research has been to engineer a good, low-loss thin-film dielectric material out of C_{60}. We have shown that, by treating the film with certain agents (molecular oxygen, nitric oxide, and chlorine), the dielectric properties of C_{60} films can indeed be improved. This may lead to potentially technologically important uses of C_{60}-derived materials in thin film electronic applications.

Much of this work was devoted to the development of scientific background for understanding the mechanism of diffusion of oxygen and other species through the bulk of the fullerite. This enhanced understanding may also prove to be technologically important, as suitable fullerene structures may provide the means to selectively separate gas mixtures. For identical time duration, pressure, and temperature conditions, while significant quantities of oxygen or NO can be incorporated into the lattice, there is no evidence that either methane [244] or nitrogen (N_2) [252] gases diffuse into the lattice. The selectivity is controlled by the channel size between interstitial sites. The channel size is, in turn, precisely determined by the fullerene size and crystal structure. Thus, extremely high levels of selectivity may be expected if the effective channel size is intermediate to the diameters of the molecules being separated. The separation process could be based on the selectivity of fullerenes to gas sorption, or, if the fullerenes are formed in a membrane [67], could be based on their selectivity to gas permeation.

A related possible application is using fullerene materials as gas adsorbants for gas storage, as well as for controlled gas release over time. Finally, with increasing interest in using fullerenes in advanced batteries, low frequency AC measurements of fullerene materials are becoming increasingly important.

5.3 Future Work

As mentioned in Sec. 4.7.4, a thickness dependence study with Al-C_{60}-Al trilayers is needed to further clarify the significance of electrode effects in our dielectric measurements. By varying the thickness of the C_{60} layer, it should be possible to reliably isolate bulk C_{60} properties from the interface effects. Preparations for such a study are currently under way.

Also, the hypothesis that the activation energy of the dielectric loss peak frequency E_τ is equal to the activation energy of the DC conductivity E_σ for C_{60} at $T > T_{01}$ (Sec. 4.8.1) needs further confirmation. More temperature dependence studies similar to that described in Sec. 4.8.1 are therefore needed.

Much of the fullerene-related work that the author has performed both at MIT and at Bell Labs is beyond the scope of this thesis. Some of the topics not mentioned here include:

- Molecular dynamics simulations of fullerene vibrational modes.
- Monte-Carlo simulation of oxygen diffusion in C_{60} lattice.
- Extensive investigations of photoconducting properties of pristine and photopolymerized C_{60}.

- Dielectric, transport, and photoconductivity measurements of C_{70}. There are indications that C_{70} may outperform C_{60} as a host material for many of the applications mentioned above. C_{70}-based compounds, therefore, merit further investigation.

- NO/C_{60} compounds looks very promising for both dielectric and microelectronics applications, and should be studied more rigorously.

- It has been observed that C_{60} interfaces with various metals may be fractal in nature. If so, the AC properties of such interfaces may exhibit interesting scaling behavior.

- Endohedral fullerenes are almost certain to have a very high dielectric constant due to their large intrinsic dipole moments. Unfortunately, the study of their bulk properties has so far been hampered by their unavailability in anything but trace amounts.

Appendix A

LabView Software Listings

The following pages contain listings of LabVIEW [314] virtual instrument (VI) drivers that we developed for the experimental configurations described in Sections 3.2.3 and 3.3. For each instrument, both the virtual instrument's front panel and the wiring diagram are presented. In some cases, the VI's position in hierarchy is also shown.

A.1 HP 4274A Impedance Bridge

See Figures A-1 through A-3.

A.2 SI 1254 Frequency Response Analyzer

See Figures A-4 and A-5.

A.3 Microdielectrometer

See Figures A-6 through A-8.

Figure A-1: HP 4274A VI's front panel.

Figure A-2: HP 4274A VI's wiring diagram. The two embedded frames of the sequence structure are shown separately.

Figure A-3: HP 4274A VI's position in hierarchy.

Figure A-4: SI 1254 VI's front panel.

Figure A-5: SI 1254 VI's wiring diagram. The six embedded frames of the sequence structure are shown separately.

Figure A-6: The Microdielectrometer VI's front panel displaying results of a representative data acquisition run.

Figure A-7: The Microdielectrometer VI's wiring diagram. The two embedded frames of the sequence structure are shown separately.

Figure A-8: The Microdielectrometer VI's position in hierarchy.

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