Transport and Dielectric Properties of Thin Fullerene (C_{60}) Films

by

Boris Pevzner

Submitted to the Department of Electrical Engineering and Computer Science
in partial fulfillment of the requirements for the degrees of

Bachelor of Science in Electrical Engineering

and

Master of Science in Electrical Engineering

at the

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Dielectric behavior of pristine and modified fullerene (C_{60}) films was investigated over the frequency range of 0.5 mHz to 100 kHz. The presence of mobile ions in the large interstitial spaces of the C_{60} lattice compromises the breakdown voltage and makes pure C_{60} unsuitable for applications requiring high-quality dielectric films (such as microelectronic applications). However, various immobile ions or neutral species (e.g., oxygen) can be made to fill the interstitial volume, changing the characteristics of the C_{60} films and, in some cases, improving their dielectric properties. Based on the low-frequency behavior of the dielectric constant, a model is developed for the mechanism for diffusion of oxygen and other impurities into the interstitial spaces of fullerene materials. It is concluded that dielectric and, especially, microdielectric spectroscopy can be used to assess the content of oxygen or other impurities in the interstitial spaces of solid C_{60}. Using this technique, both tracer and chemical diffusion coefficients can be determined, and the effect of various intercalants on the dielectric properties of C_{60} films can be ascertained.

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Preface

Since its discovery in 1985 [1], carbon-60 (C_{60}), a newly found all-carbon molecule, has captured the attention of scientists all over the world. It has been designated the 1991 "Molecule of the Year" on the cover of *Science* magazine [2], while investigation of its physical and chemical properties, as well as the properties of its compounds, became one of the most popular topics of today's research in solid state physics, material science, and chemistry.

The principal contribution of the present work has been the development of a comprehensive model for the dielectric properties of solid C_{60}, particularly explaining how the dielectric properties are affected by various dopants, impurities, and processing. The need for better dielectrics with improved properties suitable for modern integrated manufacturing needs was one of the main motivations for studying ways to enhance the dielectric properties of C_{60} thin films. And since in today's silicon technology, the enhancement of SiO_2 by nitrogen incorporation has been shown to result in an increased dielectric constant and resistance to impurity diffusion, as well as much higher resistance to interface state generation under avalanche, high-field and hot-carrier stress, and irradiation, a similar path has been chosen for attempting to improve the dielectric properties of C_{60} thin films.

It is known that solid C_{60} has a substantial amount of interstitial volume. The presence of mobile ions in these spaces compromises the breakdown voltage and makes pure C_{60} unsuitable for applications requiring high-quality dielectric films. However, various immobile ions or neutral species (e.g. oxygen) can be made to fill the interstitial volume, changing the characteristics of the C_{60} films and, in some cases, improving the dielectric properties. In this study, dielectric measurements of pristine and modified C_{60} films were performed for frequencies ranging from 0.5 mHz to 100 kHz. Based on the low-frequency behavior of the dielectric constant, a model is proposed for the mechanism of oxygen diffusion into the interstitial spaces of the C_{60} solid.

The thesis is organized into five major chapters. We start by reviewing the state of the art in the science of C_{60} and its sister materials (*fullerites*) in Chapter 1. While the theoretical and experimental knowledge in this area is still being accumulated and interpreted, a rather clear overall picture of the structure and properties of fullerites has already emerged. We briefly review synthesis of C_{60}; explain its molecular, crystalline, and electronic structure; discuss its vibrational properties and doping compounds (including superconductors), and then discuss in some detail the possible applications of this new material, paying particular attention to emerging electronic applications.

While Chapter 1 is laying the groundwork for the thesis by introducing the essential structure and properties of C_{60} necessary for understanding the material's dielectric and transport properties, Chapter 2 is similarly important as it introduces the theoretical background and mathematical machinery necessary for describing dielectric properties of materials. Chapter 3 complements Chapter 2 by explaining the techniques of low frequency dielectric measurements, including the recently developed microdielectrometry technique [3], that have been used in this study. Chapter 4 builds on the preceding chapters, and presents the data on dielectric properties of C_{60} as a function of oxygen diffusion, temperature, and other material treatments. A model for the dielectric relaxation in C_{60} that takes the role of impurities into account is also developed in Chapter 4. We conclude with a summary of main results of the work in Chapter 5.

The rationalized (MKS) system of units is used throughout this work. The typesetting convention used calls for real scalar variables to be set in regular font, complex variables — in bold font, and vector variables — with a vector sign on top.