Memory Effects for Glass-like States of Solid N_2-Ar Mixtures

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Abstract

The results of high sensitivity dielectric measurements are reported for solid N_2-Ar mixtures for N_2 concentrations $49 < x(N_2) < 100$ mole %. The observations indicate the onset of memory effects for the dielectric susceptibility on thermal cycling to temperatures below 30 K. The temperature dependence of the dielectric susceptibilities in this low temperature regime are observed to be frequency dependent, even for pure N_2 samples. These observations are consistent with a departure from ergodicity associated with the trapping of the systems in small regions of a configuration space characterized by a frustration-induced rugged free energy landscape.

Key words: glass; orientational glass; dielectric susceptibility; frustration

1. Introduction

The interplay of geometric frustration and disorder in the formation of a number of different classes of glasses has been the subject of intense recent interest [1,2]. In the simplest systems, the diatomic molecular solids (N_2, H_2, O_2.....) that form orientational or quadrupolar glasses in the presence of disorder, the frustration is caused by geometrical incompatibilities between the nearest neighbor interactions and the lattice geometries. This frustration leads to macroscopic degeneracies and a variety of new glassy phenomena at low temperatures (memory effects, local random ordering, spectral diffusion...)

Solid N_2 and solid N_2-Ar mixtures represent an ideal system for studying the interplay of geometric frustration and disorder. Below a critical concentration, long range ordering of the molecular axes is not observed, and NMR studies have shown that only a local random ordering is obtained at low temperature[3]. Heat capacity measurements show the characteristic linear temperature dependence of glassy systems, and NMR relaxation studies show that only very slow orientational relaxation occurs at low temperatures. This behavior suggest that the systems become trapped in metastable states associated with a complex free energy landscape and these should be characterized by memory effects, field cooling behavior and related non-equilibrium properties[4].

Experiments have been conducted to test this hypothesis. If the system becomes trapped in a metastable state, then one can explore the effect of applied electric fields of different frequencies that measure the dielectric response of the system associated with the tunnelling between these trapped states. It also allows the study of aging effects in the presence or absence of applied electric fields. Most significantly, the tunnelling rates are expected to be of the order of msec. which is very short compared to those of other simple glassy systems, and thus accessible to experimental tests. Note that the electric field here is not the conjugate field to the molecular alignment which would be an electric field gradient. The applied electric field induces a polarization of the molecules which then acts indirectly as the probe of the quadrupolar field response.

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2. Experimental Measurements

The anisotropic susceptibilities $\varepsilon(T)$ were measured using a 3-terminal AC capacitance bridge[5]. Figure 1 shows the frequency dependence of the relative change in $\varepsilon(T)$ for pure N_2 for 4.2 <T< 37 K. The samples were initially zero-field cooled, annealed at 50 <T< 53 K for 10-12 hours in the fields indicated, cooled to 4.2 K, and measurements taken while warming. A standard 200 V/m excitation field at 1kHz was applied to obtain the data. This particular sequence is followed because the AC capacitance bridge[5] is sensitive enough only for excitation fields stronger than 200 V/m. There is a significant reduction in $\varepsilon(T)$ for frequencies in the audio frequency range for samples cooled in external fields > 20 V/m. This reduction can result from reorientations that erase the anisotropic polarizability. For T<42 K, the excitation field has no observable effect on the history of the samples[6,7]. Figure 2 shows the history of $\varepsilon(T)$ when a solid mixture (51%N_2-Ar) is cooled in the presence of an electric field of 5 kV/m at 1kHz. For this mixture, NMR studies show that a glass state forms below 9 K. The pronounced hysteresis shows that memory effects due to aging in applied fields persist until warmed to temperatures close to those where lattice changes occur for the pure system.

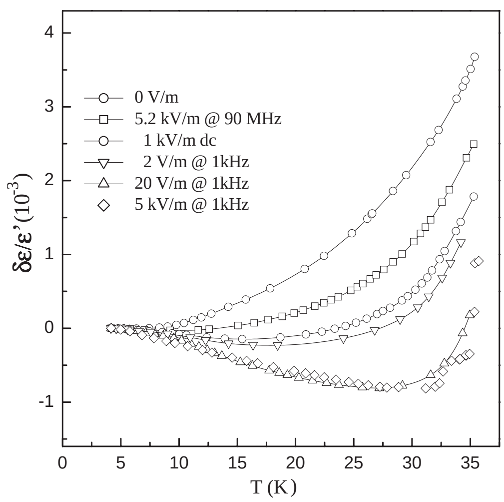

Fig. 1. Temperature dependence of the dielectric susceptibility for a range of frequencies and electric field strengths for pure N_2. $\varepsilon(T)'$ is the value at 4.2 K on each curve.

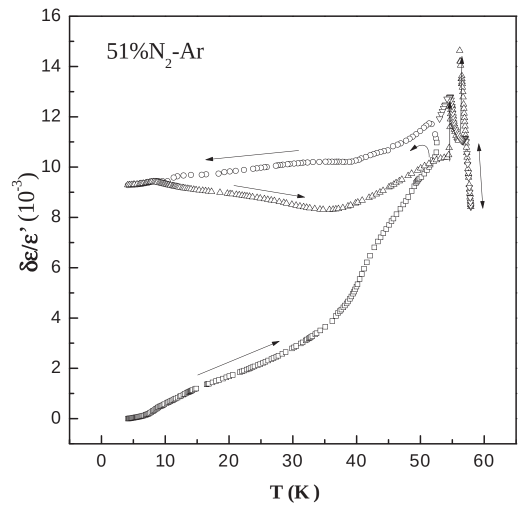

Fig. 2. Hysteresis of dielectric response for a 51%N_2-Ar solid mixture for an electric field of 1 kHz and 5 kV/m. $\varepsilon(T)'$ is the value at 4.2 K on the open squares curve.

3. Conclusions

Studies of the AC dielectric susceptibility of the N_2-Ar molecular glass former show the existence of history-dependent, field-induced nonequilibrium behaviors. The applied fields have very small perturbations on the complex energy system, but because of the dense degeneracies, they can separate states of energy differences of a few kHz and thus reduce the tunnelling rates between metastable states, leading to induced memory effects that are erased only at high temperatures. The frequency dependence for pure N_2 is not understood.

Acknowledgements

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