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Frustration-induced glass behavior in solid N_2? Audio frequency dielectric measurements

S. Pilla*, J.A. Hamida, K.A. Muttalib, N.S. Sullivan

Department of Physics, University of Florida, P.O. Box 118440, Gainesville, FL 32611-8440, USA

Abstract

High-sensitivity dielectric constant measurements of solid N_2 in the audio frequency range have revealed the existence of strong unexpected hysteresis in the high-temperature HCP phase above an onset temperature $T_h = 42$ K. We report a sharp kink at T_h, observed only when the sample is annealed at $41.5 < T < 43$ K for 6–8 h, suggesting that the thermal processes in this window of temperature are rather slow. We argue that the results are consistent with the breakdown of orientational ordering at T_h and the onset of frustration-induced orientational glass states in the absence of site disorder above T_h.

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1. Introduction

The interest in simple molecular solid N_2 is derived from the fact that this serves as a prototype for studying the simplest glass formers [1]. At low temperatures $T < T_{\alpha\beta} \approx 36$ K, a long-range periodic orientational Pa3 ordering, known as the α-phase, is observed where the FCC structure of the lattice is compatible with the ordering symmetry. The high-temperature HCP phase is incompatible with the Pa3 symmetry and the resulting orientationally disordered phase is known as the β-phase. However, the nature of this disordered phase and in particular the role of geometrical frustration in the breakdown of the long-range order has not been studied systematically.

Dielectric constant measurements in these systems are particularly important because they directly probe the orientational ordering. The relevant microscopic time scale is in the audio frequency range due to the hindered motion of the overlapping N_2 molecules [1]. We have developed a three terminal AC capacitance bridge with two parts per billion sensitivity in the 0.2–20 kHz range for measuring the real part of the dielectric constant $\varepsilon(T)$ as a function of temperature T [4].

2. Results and discussion

Recently, strong hysteresis effect has been observed in pure N_2 in the high-temperature HCP phase above an onset temperature $T_h = 42$ K [2]. For thermal cycling below T_h there is no hysteresis. In this regime $\varepsilon(T)$ retraces a unique curve depending on the initial conditions, including the jump at $T_{\alpha\beta}$. However, if the system is taken above T_h during a heating cycle (with the AC electric field present), the observed $\varepsilon(T)$ is higher than the previous curve. The difference between the two curves depends on how far above T_h the system was heated. Fig. 1 shows typical hysteresis curves for a 1 kHz electric field of strength 5 kV/m. In Fig. 1 the system was heated to 48 K along curve 1 and cooled along curve 4. When the system is heated to a temperature less than 48 K but above T_h and cooled, the $\varepsilon(T)$ lies between curves 1 and 4. In addition to these previous observations [2] we have also observed in zero-field cooled samples a sharp kink around T_h, when we annealed the sample at $41.5 < T < 43$ K for 6–8 h during the warm-up cycle.

* Corresponding author.
E-mail address: manyam@phys.ufl.edu (S. Pilla)

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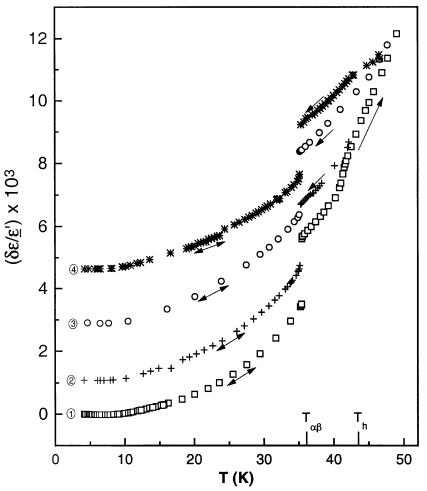

Fig. 1. $\varepsilon(T)$ of solid N_2 at 5 kV/m and 1 kHz excitation field relative to the value at 4.2 K on curve 1 ($\underline{\varepsilon}'$). Different curves show the hysteresis effects in α-phase.

But when the sample is heated through this window of temperature rather rapidly (say in $\frac{1}{2}$ h) the kink could not be observed [2], suggesting that the thermal processes in this temperature window are rather slow. Once the sample is cooled in the presence of the AC excitation field (say curve 2) and warmed again to temperatures above T_h, the kink we observed for curve 1 (the zero field cooled sample) at T_h could no longer be observed, irrespective of the annealing time at T_h. Also, the kink is absent during all cooling cycles irrespective of the annealing time at T_h. That is, the kink is present only for zero-field cooled samples during warming cycles. When the above field-cooled sample is annealed at $T > T_h$ for 12–15 h in the absence of the AC field and cooled down to 4.2 K, we always obtained curve 1. This indicates that the existence of the strong hysteresis above T_h for pure N_2 cannot be described by postulating the existence of mobile vacancies or dislocations. In the absence of such mobile defects, the strong hysteresis signals the existence of glass-like states. We note that thermal resistivity data of pure N_2 [3] also shows an anomaly close to T_h. Because the anisotropic polarizability [2] probes the local correlations between the orientational order parameters and the orientations of the molecular axes, the observed hysteresis indicates that these correlations are glass-like in the sense that the system becomes trapped in a different part of the phase space depending on the thermal history.

We note that there are disorder-induced orientational glass states for $T < 25$ K in the N_2–Ar mixtures [1]. In the absence of site disorder in case of pure N_2, the hysteresis must be induced by a competition between the orientational ordering and the geometrical frustration. In particular, the thermal processes in N_2 near T_h are slow, of the order of several hours, making N_2 an ideal system for studying non-equilibrium effects because the time scales involved are accessible to experiment with high sensitivity.

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