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Orientational ordering of solid CO: high sensitivity dielectric measurements

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Abstract

Dielectric constant measurements of solid N_2 have revealed strong hysteresis effects due to geometrical frustration. To test the universality of such glassy behavior arising purely from geometrical frustration, we have made dielectric constant/loss measurements on a similar system, CO. We find that CO does not show any hysteresis effects in the entire solid phase. We suspect that this is due either to the very small window of temperature where CO exists in its orientationally disordered solid phase or to the presence of a small intrinsic dipole moment making it different from a pure quadrupolar system such as N_2.

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1. Introduction

Frustration plays an important role in the formation of glasses in a wide variety of systems. Pure N_2 and CO with their intrinsic electric quadrupole moments are model systems for that purpose [1,2]. In these systems frustration arises from a basic incompatibility between the symmetry properties of the quadrupole–quadrupole interactions and the lattice geometry. In the high-temperature solid phase both N_2 and CO form HCP lattices which cannot support an orientationally ordered Pa3 structure which is favored by the quadrupolar interactions. At low temperatures the crystal structure for both changes from HCP to FCC, which does support the Pa3 structure, and the molecules form an orientationally ordered state. Because the anisotropic part of the polarizability tensor probes the orientational states of the molecule, systematic studies of the dielectric constant/loss and their temperature dependencies will be able to differentiate amongst orientationally ordered, disordered, and glassy states.

2. Experimental

Recent dielectric measurements on pure N_2 [1] have revealed surprisingly strong hysteresis effects in the high T HCP phase due to geometrical frustration. In order to test the universality of such glassy behavior, we have made systematic measurements on CO, which also has a small intrinsic dipole moment which makes it more accessible to dielectric studies. To carry out our measurements we have built a low noise three-terminal AC capacitance bridge with a sensitivity of two parts per billion for measuring dielectric constant ε as a function of temperature in 0.2–20 kHz frequency range [3].

3. Results and discussion

Fig. 1 shows the warming and cooling data for $\varepsilon(T)$ of CO at 1 kHz. For $T < 25$ K, $\varepsilon(T)$ for polar CO initially decreases with T (not shown in Fig. 1) and then increases with a dip in $\varepsilon(T)$ near 7.5 K similar to N_2 [1]. This shows there is no long-range ordering of the dipoles down to 4.2 K. T_P therefore signals the onset of dynamical freezing of the dipoles in random orientations. At T_P the dipoles flipped by thermal excitations contribute

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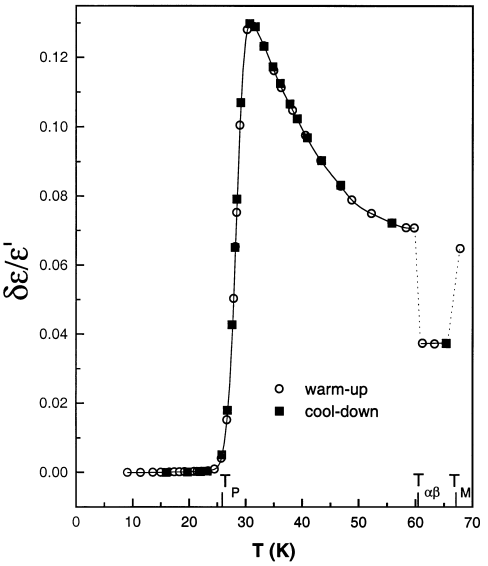

Fig. 1. Dielectric constant of solid CO at 1 kHz relative to the value of ε at 4.2 K (ε'). No hysteresis was observed in the entire temperature range studied.

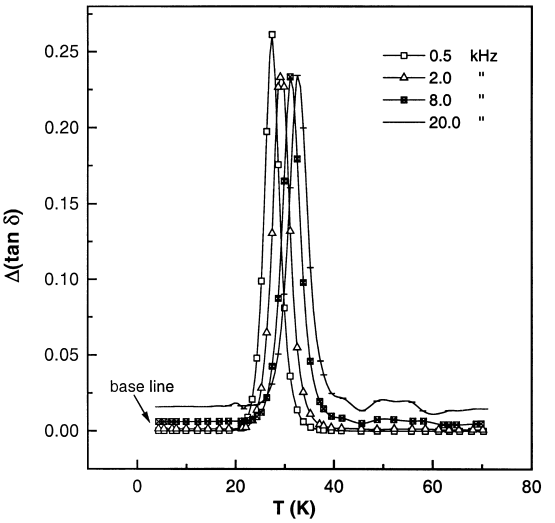

Fig. 2. Variation of the loss angle with temperature for solid CO.

significantly to ε. A large jump in $\varepsilon(T)$ is observed at the first-order transition from FCC to HCP phase at $T_{\alpha\beta}$. These high-resolution results show that there is no hysteresis in the entire solid phase. This is in contrast to the strong hysteresis effect observed for solid N_2 in its orientationally disordered β phase [1]. This may be due to the presence of the small intrinsic dipole moment of CO. However, we suspect that the difference may also be due to the small window of temperature $T_{\alpha\beta} < T < T_M$ ($T_{\alpha\beta}$ is the structural transition temperature and T_M is the melting temperature), where CO exists in the orientationally disordered β phase. It is possible that the onset of hysteresis in CO occurs too close to T_M, which prevents us from observing it. Fig. 2 shows the variation of the loss angle $\Delta(\tan\delta)$ for CO. The temperatures for which the loss angle has a maximum match very well with the dipolar freezing temperature T_P [1] where ε(T) also has peaks. Fig. 2 indicates that the loss peaks level off for frequencies above 2 kHz. The loss angle for pure N_2 is independent of temperature for all frequencies studied (not shown).

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