

## Hemberger et al. reply

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**Hemberger *et al.* Reply:** Our Letter [1] presented evidence for a static freezing near  $T_f = 30$  K in betaine orientational glasses (OG) using several independent experimental techniques. Kim and Kim [2] challenge the interpretation of our data mainly because the frequency dependence of the linear dielectric constant of D-BP:BPI (40:60) persists below  $T_f$ . In this Reply we show that several of their comments are misleading or have been addressed already in the original paper. We maintain that the analysis and the interpretation presented in [1] are correct in terms of the present understanding of the OG transition.

In order to clarify the first two questions by Kim and Kim we reiterate from [1] that only the *slowest* element in the relaxation time spectrum  $g(\tau)$  is associated with the static freezing. This element corresponds to  $\delta \rightarrow 1$  in the representation introduced by Kutnjak *et al.* [3]. The symmetric Gaussian approximation on which the reasoning in [2] is based is clearly inappropriate for the description of  $g(\tau)$  in D-BP:BPI (40:60), since the dielectric loss curves of this compound (cf. Fig. 1 in [1]), like those of other OG [3], are strongly asymmetric. Furthermore it is well documented also for other OG [3] that even thermally activated hopping processes can persist well below  $T_f$  for elements corresponding to  $\delta < 1$ . This means that except for  $\delta = 1$  one always has Vogel temperatures  $T_0 < T_f$ .

The third issue raised by Kim and Kim concerns the rate dependence of the FH/ZFH susceptibilities which, as pointed out in [1], should exist. The only systematic study of such a dependence in OG that we know of [4] did not yield evidence for a significant rate dependence of  $T_f$ . This suggests that even if such a dependence existed in OG, it must be extremely weak. In any case the experimental time scale of about  $10^3$  s associated with a rate of 1 K/min [5] is much slower than the characteristic relaxation time ( $\tau \approx 1$  s) near 30 K, thus providing quasistatic conditions for our measurements [6].

Kim and Kim's final remark concerns the nonlinear susceptibility  $\chi_{NL}$ . In disordered systems this quantity usually only exhibits dispersion in temperature ranges for which also the corresponding linear susceptibilities are frequency dependent [7]. Since this fact may not be widely known, we have explicitly stated in [1] that our data analysis was restricted to a temperature range where no dispersion in  $\chi_1$  and  $\chi_3$  occurs for the selected measuring frequency  $\nu$ .

Of course the dispersion of  $\chi_{NL}$  (near  $T_f$ ) is interesting in itself. In Fig. 1 we present the frequency dependence of the imaginary part of the third order susceptibility, denoted as  $\chi_3''$ , for several temperatures. As is the case for the absorptive part of the linear susceptibility,  $\chi_1''(\nu)$ , indications for a static freezing transition are not obvious from  $\chi_3''(\nu)$  as well, at first glance. The inset of Fig. 1 shows that the minima of  $\chi_3''$  exhibit a similar temperature dependence than the maxima of  $\chi_1''$ , taken from [1]. This result, again, provides clear experimental evidence that an analysis in terms of average, principal, most contributing,

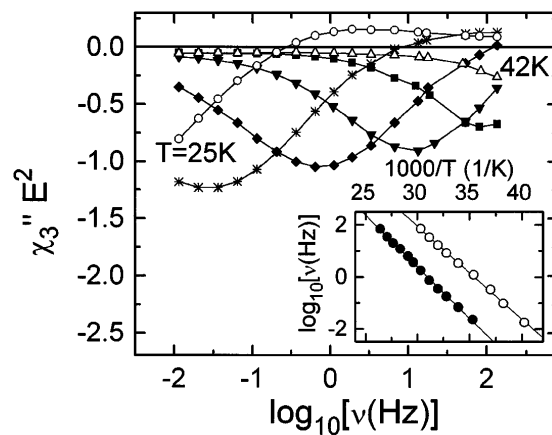


FIG. 1. Spectra of the imaginary part of the third order susceptibility of D-BP:BPI (40:60) as measured for the following temperatures: 25, 27, 31, 35, 40, and 42 K. The data were taken using ac fields of up to 1100 V/mm. The inset shows an Arrhenius representation of the minima of these spectra (closed symbols) and of the maxima of  $\chi_1''$  as taken from [1] (open symbols).

etc. relaxation times, even those of higher order, do not yield any valuable insights into the static freezing transition in betaine OG. In conclusion, Kim and Kim raise the question whether relaxation of orientational degrees of freedom is possible below a quasistatic freezing temperature. Our experimental results demonstrate that the answer is affirmative, at least as long as the Edwards-Anderson order parameter at  $T_f$  is smaller than unity. This condition is, however, met for all known OG.

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