Erratum: From a Simple Liquid to a Polymer Melt: NMR Relaxometry Study of Polybutadiene [Phys. Rev. Lett. 97, 207803 (2006)]

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In our recent publication [1], we analyzed the NMR field cycling spectra of a series of polybutadienes (PB) of molecular weights (in g/mol) from $M = 355$ to $M = 817000$. The spectra were decomposed in "polymer spectra" and "glass spectra." With increasing M, the weight of the polymer spectrum increases at the expense of that of the glass spectrum. As a measure of the spectral weight of the polymer contribution, we introduced the quantity S. The polymer spectra were then analyzed within Rouse theory. As short polymer chains were involved, the discreet Rouse model has to be applied. Here, a numerical error appeared leading to Rouse spectra being correct in spectral shape but not correctly normalized. The correct spectra as a function of number of Rouse segments N are shown in Fig. 1(a). Independent of N, the spectra exhibit the same integral but exhibit successively more intensity at low frequencies. Correspondingly, formula (5a) in [1] is incorrect; i.e., the dependence of S on N cannot be explained by standard Rouse theory. In order to compare the result of Fig. 1(a) with the experimental polymer spectra, one also has to normalize the latter. Hence, in Fig. 1(b), we show the polymer spectra normalized by their weights S. Now the prediction of Rouse theory regarding the spectral density at zero frequency, explicitly $J(0) \propto \ln N$, can be tested. The insert in Fig. 1(a) displays the result. Introducing the Rouse unit $M_R = 500$ and $M = NM_R$, as before [1], only the three PB samples with $777 \leq M \leq 2020$ behave according to the prediction of the theory. Already at $M > 2020$, indications of entanglement dynamics show up. Thus, the results of our study remain unchanged.

A further comment is worthwhile. The quantity S was called the "order parameter" [1]. Here, certain confusion exists in the literature as to whether this quantity is identified with the order parameter itself, or S denotes the square of the order parameter. Following the well established meaning of the order parameter in the field of liquid crystals, the introduced quantity S represents the square of the order parameter [2].

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[1] S. Kariyo, C. Gainaru, H. Schick, A. Brodin, V. N. Novikov, and E. A. Rössler, Phys. Rev. Lett. **97**, 207803 (2006).
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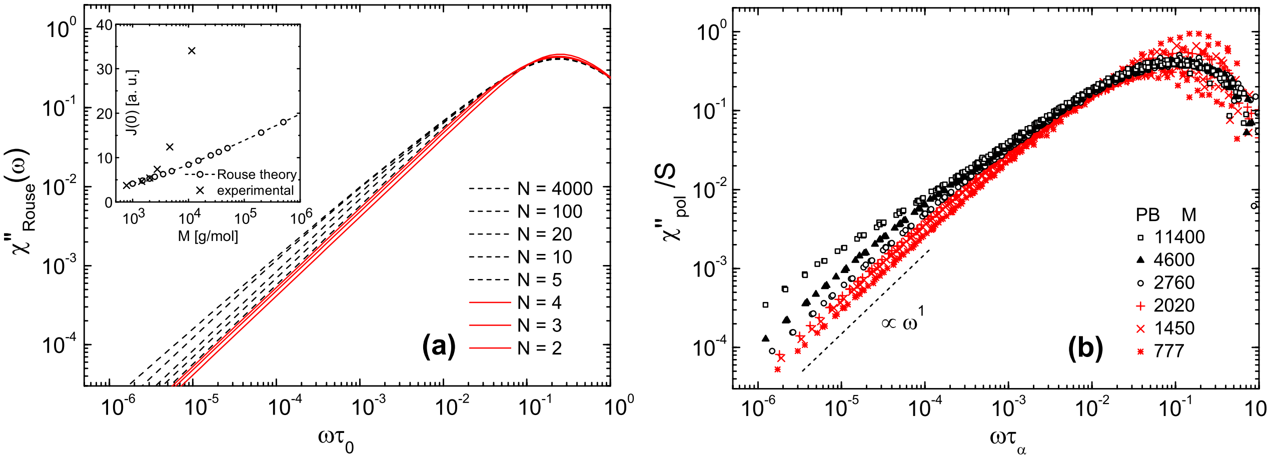

FIG. 1 (color online). (a) Susceptibility spectra for different number N of beads as calculated from Rouse theory. Insert: Comparison of the low frequency limit $J(0)$ as obtained from Rouse theory (circles) and experimentally (crosses). A molecular weight $M_R = 500$ of the Rouse unit is introduced, and the experimental data are scaled. (b) Normalized experimental polymer spectra for different molecular weights. Rouse theory applies for $M \leq 2020$; at larger M, entanglement effects set in.