

and the temperature dependence of  $T_1$  and  $T_2$  data available for glycerol, BUT and MPD. Reduction of both types of data, for glycerol, give identical results for temperatures above 12°C with some discrepancy at lower temperatures. The recovery of the self-diffusion constant at atmospheric pressure supports the use of the temperature reduction.

Comparison of  $1/T_{1\text{rot}}$  in perdeuterated glycerol to  $1/T_1$  data indicates that the rotational contribution must be less than 10% in the region of the  $T_1$  minimum to allow a reasonable fit to the translational part of  $1/T_1$ .

The above results represent a reversal of previous notions concerning the strength of rotational and translational relaxation processes in viscous liquids and underline the importance of considering translational diffusive effects.

$\ln\tau_i$  vs  $T$  is found to be non-Arrhenius for all three liquids.  $\ln\tau_i$  vs  $P$  is linear in glycerol.  $\ln D$  vs  $P$  is linear in glycerol and BUT. In MPD and glycerol, for which  $\eta_s(P)$  has been measured,  $D$  is not proportional to  $\eta_s^{-1}$ .

A good fit to both temperature and pressure NMR data is possible with fixed values of  $\alpha$  for each of the three liquids studied through the complete temperature and pressure range of the measurements.

The fitted values of  $d$  are less than a molecular diameter. However, if  $d$  is considered a measure of the closest distance between protons on neighboring molecules, the magnitudes obtained are not unreasonable.

The effects of off-center spins and a nonuniform distribution of intermolecular spins in accordance with the radial distribution functions is found to be negligible (at least at low  $\omega\tau$ ) in  $1/T_{1\text{inter}}$  for glycerol.

Comparison of  $\tau_i$  and  $\tau_j$  with other times available in literature, does not indicate any apparent relationships though the temperature dependences of all  $\tau$ 's are similar. This may indicate that motions other than translational motions may be involved in the processes measured by these times.

#### ACKNOWLEDGMENT

The authors would like to thank P. W. Drake very much for his continued assistance during the experiment and for his very careful and critical reading of the manuscript.

\* This work has been sponsored by the National Science Foundation, No. GP-9410.

<sup>1</sup> N. Bloembergen, E. M. Purcell, and R. V. Pound, Phys. Rev. **73**, 679 (1948).

<sup>2</sup> B. I. Hunt, J. G. Powles, Proc. Phys. Soc. **88**, 513 (1966).

<sup>3</sup> S. H. Glarum, J. Chem. Phys. **33**, 639 (1960).

<sup>4</sup> S. H. Glarum, J. Chem. Phys. **33**, 1371 (1960).

<sup>5</sup> I. Chaban, Zh. Eksp. Teor. Fiz. **53**, 556 (1968) [Sov. Phys. JETP **26**, 361 (1968)].

<sup>6</sup> M. A. Isakovich, I. A. Chaban, Zh. Eksp. Teor. Fiz. **50**, 1343 (1966) [Sov. Phys. JETP **23**, 893 (1966)].

<sup>7</sup> F. Noack, G. Preissing, Z. Naturforsch. **24a**, 143 (1969).

<sup>8</sup> A. Favret, R. Meister, J. Chem. Phys. **41**, 1011 (1964).

<sup>9</sup> F. Noack, G. Preissing, in *Magnetic Resonance and Relaxation*, Proc. Colloque Ampere, 14th, Ljublyana, 1966 (North-Holland, Amsterdam, 1967).

<sup>10</sup> H. C. Torrey, Phys. Rev. **92**, 962 (1953).

<sup>11</sup> NP have recently indicated in a brief note a revised view of this interpretation in view of measurements on the self-diffusion constant [Verhandl. Deut. Phys. Ges. (VI), **5**, 347 (1970)]. A further analysis of glycerol based on the translational model which parallels our discussion has also been done [Z. Physik, **246**, 84 (1971)].

<sup>12</sup> G. J. Kruger, Z. Naturforsch. **24a**, 560 (1969).

<sup>13</sup> G. Benedek, E. M. Purcell, J. Chem. Phys. **22**, 2003 (1954).

<sup>14</sup> G. Benedek, Ph.D. thesis, Harvard University 1953, (unpublished).

<sup>15</sup> T. E. Bull, J. Jonas, J. Chem. Phys. **52**, 4553 (1970).

<sup>16</sup> R. Kubo and K. Tomita, J. Phys. Soc. Japan **9**, 316 (1954).

<sup>17</sup> J. Harmon and B. Muller, Phys. Rev. **182**, 400 (1969).

<sup>18</sup> P. Hubbard, Phys. Rev. **131**, 275 (1963).

<sup>19</sup> R. Kubo, in *Fluctuation, Relaxation, and Resonance in Magnetic Systems*, edited by D. ter Haar (Oliver and Boyd, London, 1962).

<sup>20</sup> See also, F. Noack and G. Held, Z. Physik. **210**, 60 (1968) for a discussion of  $\tau_c$  and  $K(\tau)$  for nonexponential relaxation functions.

<sup>21</sup> See, E. G. Abragam, *The Principles of Nuclear Magnetism* (Oxford U. P., Oxford, 1961), pp. 297-302.

<sup>22</sup> A. A. Brook, E. O. Stejskal, and V. W. Weiss, Rev. Sci. Instr. **39**, 917 (1968).

<sup>23</sup> The author wishes to thank and acknowledge Dr. E. O. Stejskal who provided the machine drawings for this cell and a sample of the teflon material used.

<sup>24</sup> A. Favret, and R. Meister, Rev. Sci. Instr. **36**, 154 (1965).

<sup>25</sup> C. Moynihan and P. Drake (private communication).

<sup>26</sup> The viscosity data was checked directly with the data of: L. E. Cotter, (M. S. thesis, Catholic University, Wash., D.C.) for 1,3-butanediol; R. J. Sciamanda (M.S. thesis, C.U., Wash., D.C.) and G. McDuffie (private communication), for 2-methyl-2,4-pentanediol and found to be in good agreement.

<sup>27</sup> T. Connor, Trans. Faraday Soc. **60**, 1574 (1964).

<sup>28</sup> K. Kuszczynski, J. Kail, and J. Powles, Proc. Phys. Soc. **75**, 243 (1960).

<sup>29</sup> P. Drake (private communication).

<sup>30</sup> The diffusion data reported here is taken from: P. Gammell (Ph. D. thesis, C. U., Wash., D. C., 1971) and private communication (glycerol, MPD, BUT), E. O. Stejskal, and J. Tanner, J. Chem. Phys. **42**, 288 (1965) (glycerol), F. Noack and G. Preissing, Verhandl. Deut. Phys. Ges. (VI), **5**, 347 (1970) (glycerol).

<sup>31</sup> See Ref. 9 Eq. (4a).

<sup>32</sup> The predictions for MPD based on the 30 MHz fit failed to agree with the 10 and 20 MHz data of Ref. 11. The MPD data was rerun at 20 MHz and was found to be in good agreement with the predictions.

<sup>33</sup> P. Drake and R. Meister, J. Chem. Phys. **54**, 3046 (1971).

<sup>34</sup> See Ref. 33 Eq. (2) and (3).

<sup>35</sup> See Ref. 21, p. 314.

<sup>36</sup>  $\tau_{or}$ ,  $\tau_s$  and  $\tau_{Diel}$  for glycerol were taken from Ref. 33.

<sup>37</sup>  $\tau_p$  and  $\tau_v$  were calculated from viscosity data mentioned in Ref. 26. The following empirical formulas for  $\eta_s$  were used to calculate  $\tau_v$  in glycerol and  $\tau_s$ ,  $\tau_v$ ,  $\tau_p$  in BUT and MPD.

$$\text{MPD } \eta_s = \exp[-3.315 + 1.74 \times 10^8 / (T + 273)^2]$$

$$\text{BUT } \eta_s = \exp[-1.3776 + 1.5744 \times 10^8 / (T + 273)^2]$$

$$\text{GLY } \eta_s = \exp[2.30258[-0.724 + 0.98 \times 10^8 / (T + 273)^2]]$$

The Moduli data used is taken from R. Meister *et al.*, J. Acoustical. Soc. **31**, 854 (1960).

<sup>38</sup> J. Harmon, Chem. Phys. Letters **7**, 207 (1970).

<sup>39</sup> J. Tauke, M. A. thesis, Physics Dept., C. U., Wash., D. C., (unpublished).

<sup>40</sup> W. Slie and W. Madigosky, J. Chem. Phys. **48**, 2810 (1968).

<sup>41</sup> G. McDuffie and M. Kelly, J. Chem. Phys. **41**, 2666 (1964).