

corresponding values are 5.8 and 67.7 for NH_4Cl .³ In the case of the c' wave, the relaxation strength is less well established and it does not seem justifiable to attempt an analytical representation.

The relaxation times obtained from Eq. (4) by using the experimental α_c/ω^2 data in Table I and the above choices of C values are shown in Fig. 7, where $1/\tau$ is plotted versus ΔT . This figure indicates that the longitudinal relaxation times can be well represented by

$$1/\tau = 1/\tau(0) + a\epsilon^n, \quad (7)$$

where the subscript S, x has been dropped for convenience. The parameters $1/\tau(0)$ and a corresponding to the line in Fig. 7 are 1.8 and 1780 (in units of 10^8 sec^{-1}) and the exponent $n=1$. However, the limiting value of τ at T_λ should not be considered as well established, since a reasonably good fit can also be achieved with $1/\tau(0)=0$ and $a=1830$. The transverse relaxation times are quite similar in magnitude to the longitudinal τ values but appear to vary in a systematically different manner. Indeed, the best fit to these $1/\tau$ values gives an exponent $n=1.25$. A decision as to whether such differences are real or not will require reliable values of c'' as a function of temperature.

It is important to note that the fit to the attenuation using Eq. (4) with experimental C values and the form for τ given in Eq. (7) is much better throughout the entire temperature range than the fit with Eq. (1). The χ^2 value is quite good (0.9) and there are no systematic deviations. Even if a single relaxation formalism is not valid, the analysis we have carried out for low-frequency attenuation data is an appropriate way to treat the slowly varying strength C and obtain the exponent n for the critical relaxation frequency. That is, we can reinterpret Eq. (4) as the low-frequency form of a more general relaxation theory involving a spectrum of relaxation times. If the sound wave is coupled to the order-parameter fluctuations via a dependence of the NH_4^+ -ion interactions on the lattice parameter (volume magnetostrictive coupling in the analogous magnetic case), the major contributions to α at small ω should come from fluctuations with $k \approx \kappa$, where κ is the inverse correlation length.¹⁴ In this case, our $1/\tau \propto \Omega_\kappa$,

which is the characteristic frequency for order-parameter fluctuations with wave vector κ . Since the ammonium halides are formally quite similar to magnetic systems in their ordering, we will briefly state a few theoretical results for antiferromagnets. For an isotropic Heisenberg antiferromagnet¹⁶ n would equal $\theta\nu$, where $\theta = \frac{2}{3}$ and ν is the exponent for the inverse correlation length ($\kappa = \kappa_0 \epsilon^\nu$). Thus, the conventional choice of $\nu = \frac{2}{3}$ would be consistent with our value of $n=1$. However, the Hamiltonian for NH_4Br is complicated owing to the presence of two competing interactions,² and it does not seem that one should expect agreement with theories based on an isotropic Heisenberg model. The behavior of the critical spin relaxation rate for an anisotropic Heisenberg antiferromagnetic seems to be best described by "conventional" (van Hove-type) theory,^{17,18} according to which $1/\tau$ should vary like the inverse susceptibility. Thus the exponent n should equal γ , which on the basis of scaling relations is given by $\gamma = (2-\eta)\nu$. The usual choice of $\eta=0$ and $\nu \approx \frac{2}{3}$ gives $\gamma \approx \frac{4}{3}$, in poor agreement with n . In much previous acoustic work, the variation of the relaxation strength was unknown and ignored, which led to the assumption that the exponent l in Eq. (1) was the same as n . In this view, the value $l=1.35$ would appear to be consistent with $\gamma \approx \frac{4}{3}$, but we consider this to be a fortuitous agreement. Indeed, if the conventional magnetic theory is appropriate to NH_4Br , the value of n would be consistent with the classical (Landau) exponents $\eta=0$, $\nu=\frac{1}{2}$, $\gamma=1$. Further progress requires a better understanding of the effective Hamiltonian for NH_4Br and greater development of the dynamical theories of critical phenomena.

As a final point, let us compare the dynamical aspects of the acoustic behavior of NH_4Br with those of NH_4Cl . Although the behavior of the maximum dispersion (i.e., the longitudinal relaxation strength C) is very similar in the disordered phase of NH_4Cl and NH_4Br , the variation in the relaxation time is strikingly different. For NH_4Cl at 1 atm $1/\tau = 414 \times 10^8 \epsilon^{0.6} \text{ sec}^{-1}$ up to $\Delta T = +50^\circ \text{K}$.³ Furthermore, there is no dispersion or critical attenuation associated with the c' wave in NH_4Cl . Thus the dynamical critical behavior in these two isomorphous crystals differs considerably.

¹Work supported in part by the National Science Foundation.

²C. W. Garland and R. A. Young, *J. Chem. Phys.* **49**, 5282 (1968).

³Y. Yamada, M. Mori, and Y. Noda, *J. Phys. Soc. Jap.* **32**, 1565 (1972); A. Hüller, *Z. Phys.* **254**, 456 (1972).

⁴C. W. Garland and R. J. Pollina, *J. Chem. Phys.* **58**, 5002 (1973).

⁵C. W. Garland and C. F. Yarnell, *J. Chem. Phys.* **44**, 1112 (1966).

⁶G. J. Rosasco, C. Benoit, and A. Weber, in *Proceedings of the Second International Conference on Light Scattering of Solids*, edited by M. Bakanski (Flammarion, Paris, 1971), p. 483.

⁷C. W. Garland and D. D. Snyder, *J. Phys. Chem. Solids* **31**, 1759 (1970).

⁸C. W. Garland and R. Renard, *J. Chem. Phys.* **44**, 1130 (1966).

⁹C. K. Choo and C. W. Garland, *J. Chem. Phys.* **59**, 1541 (1973).

¹⁰According to C. H. Wang and R. B. Wright [*J. Chem. Phys.*]