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Consideration of a Frequency-Independent Damping Coefficient from Raman Measurements*

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The possibility of a frequency-independent or frequency-dependent damping constant, γ, is experimentally investigated, using the right-angle Raman results for the heavily damped soft-E mode in the ferroelectric $PbTiO_3$ at 491°C (just below the transition temperature). The fits, over a wide frequency range, using a frequency-independent γ are so good that there is little room for any appreciable frequency dependence. This is in disagreement with several claims made in a recent paper by Heiman, Ushioda, and Remeika.

In a recent paper Heiman, Ushioda, and Remeika[1] (HUR) report polariton measurements of the soft-E mode in $PbTiO_3$. From these measurements they determine, at room temperature ≈ 20°C, the frequency dependence of the damping constant $\gamma(\omega, T)$ for 45 cm$^{-1} < \omega <$ 90 cm^{-1} shown in Fig. 1. Previous to the polariton measurements, Burns and Scott[2] (BS) have performed right-angle Raman measurements on this same mode in $PbTiO_3$ as a function of temperature up to the transition temperature $T_c = 493$°C. BS fitted the line shape between 20 cm$^{-1} < \omega <$ 100 cm^{-1} using a frequency-independent damping constant, $\gamma(T)$, and showed that $\gamma(T)$ diverges as T approaches T_c from below.

Using their results, HUR claim[1] that the divergence of the damping constant as $T \to T_c$ arises mainly from the frequency dependence as measured at 20°C and thus there is no singular behavior in $\gamma(T)$, and claim that $\gamma(\omega, T)$, at a fixed frequency, is proportional to absolute temperature. I investigate these claims.

The right-angle Raman results[2] may be fitted[3] with a simple-harmonic-oscillator response function with velocity damping ($\ddot{x} + \gamma\dot{x} + \omega_0^2 x = E$) which for the intensity I of the Raman-Stokes line yields

$$I = A(1+n)\omega\gamma\omega_0^2/[(\omega_0^2 - \omega^2)^2 + \omega^2\gamma^2], \qquad (1)$$

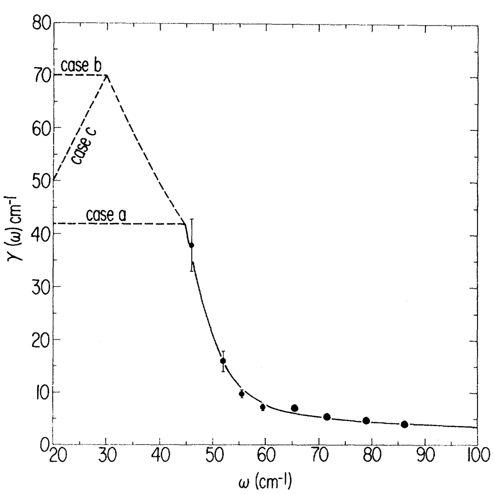

FIG. 1. Damping coefficient versus frequency for $PbTiO_3$. The solid circles are the room-temperature results of HUR. The solid line, from 45 to 100 cm^{-1}, is used for $\gamma(\omega, T = 20°C)$. Three different extrapolations are shown. These are used for the 20–100-cm^{-1} data.

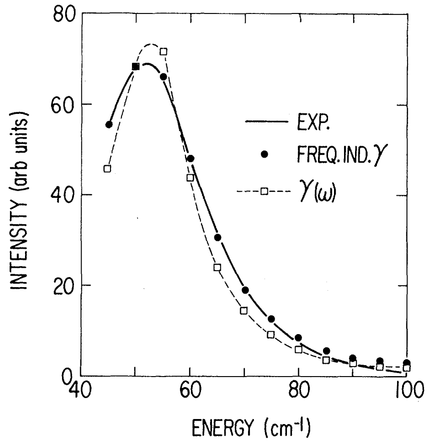

FIG. 2. A comparison of fits, using a frequency-independent and a frequency-dependent γ, to the experimental right-angle Raman data for the soft-E mode in $PbTiO_3$ at 491°C. No extrapolation of the HUR data is required in this range of ω.

where A is a frequency-independent constant, n is the Bose-Einstein factor $[\exp(\hbar\omega/kT)-1]^{-1}$, ω_0 is the harmonic frequency, and γ is the damping constant. We take HUR's result for $\gamma(\omega, T=20°C)$ for 45 $cm^{-1} < \omega < 90$ cm^{-1} shown in Fig. 1, correct it for temperature [since it is claimed to be proportional to temperature we multiply it by $(491+273)/293$], and fit to the 491°C experimental result. A least-squares fit is performed to determine the best value of ω_0 in Eq. (1). One may also fit to the experimental data with a frequency independent γ and determine, also by a least-squares fit, the best value of ω_0 and γ. The results for both of these fits in this limited frequency range is shown in Fig. 2. The quality of the fit can be indicated by the mean square deviation (MSD)

$$(1/N)\sum_{i=1}^{N}(D_i - C_i)^2, \qquad (2)$$

where D_i are the data points, and C_i are the corresponding points calculated for the best fit. As can be seen in Fig. 2, the result using a frequency-independent damping constant is excellent and probably within the limit of error of the experimental data.[2] The fit using HUR's frequency-dependent damping is not good, especially around the peak of the experimental data. Table I summarizes the results in the 45- to 100-cm^{-1} range and includes the MSD values.

If the two different fits, shown in Fig. 2, were all that could be said about the different damping constants perhaps the results would not be very conclusive. However, the data taken at[2] 491°C extend over a wider frequency range then the polariton data.[1] HUR make a strong proposal that their $\gamma(\omega, T=20°C)$ result will peak at ≈ 30 cm^{-1}. In Fig. 1 I show three possible cases of an extrapolation of the HUR result. Case c is their proposal. As for the previous frequency range, one may fit Eq. (1) to the experimental data using a frequency-independent damping as well as using $\gamma(\omega, T=20°C)$ extrapolated as shown in Fig. 1 and multiplied by $(491+273)/293$ as claimed by HUR. The results of the fits are shown in Table I. Figure 3 shows the experimental data and the results of the fit using a frequency-independent damping as well as the result from case b

TABLE I. Results from fitting to the 491°C right-angle Raman data for $PbTiO_3$.

Data (cm^{-1})	ω_0 (cm^{-1})	γ (cm^{-1})	MSD
45–100	54.86	24.95	1.29
45–100	42.20	HUR	23.0
20–100	54.88	24.82	1.18
20–100	52.80	case a	1050
20–100	49.78	case b	661
20–100	51.35	case c	814

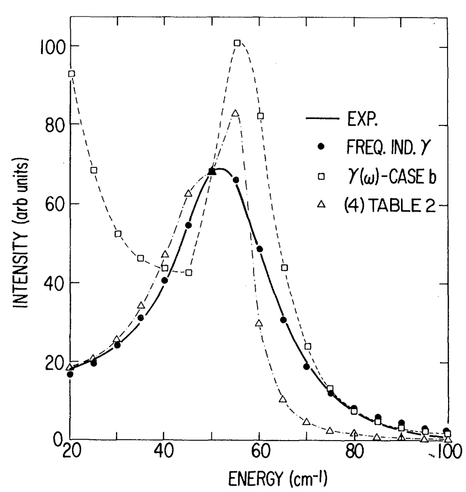

FIG. 3. The same as Fig. 2 except that the HUR data are extrapolated to 20 cm^{-1}. Case b (the squares) is shown because it gives the lowest MSD. The least-squares results for cases a and c are in general very similar to case b except a little poorer. For example the point at 20 cm^{-1} occurs at 98 and 103 for a and c, respectively, compared to 92 for b. The triangles show the result for (4) from Table II which is the best of the two-parameter fits where the lowest frequency point of the polariton data is de-emphasized.

which has the smallest MSD of the three cases. As can be seen, any of the extrapolated results for the frequency-dependent damping are very poor.

Since the lowest-frequency datum point of HUR has the largest uncertainty (see Fig. 1), we would like to try fitting the 491°C data with less emphasis on this isolated point. One way to do this is as follows: The lower limit of this datum point is $\gamma = 32.8$ cm^{-1}. So we fit the experimental data in exactly the same manner as for the result in Fig. 2, but for $\omega = 45$ cm^{-1} we take $\gamma = 32.8$ cm^{-1} instead of $\gamma = 42$ cm^{-1}. The result is $\omega_0 = 42.84$ cm^{-1} and the MSD is 15.8. The principal difference between this result and that shown in Fig. 2 is that this result fits much better at $\omega = 45$ cm^{-1} and a bit worse at[4] $\omega = 55$ cm^{-1}. To eliminate the last point altogether the data were analyzed in the same way but, from the curve in Fig. 1, the lowest frequency point was taken as $\gamma(\omega) = 21.5$ cm^{-1} at $\omega = 50$ cm^{-1}. The result of the fit is $\omega_0 = 42.69$ cm^{-1} and a MSD of 16.6, where the fit is quite similar to that shown in Fig. 2 for $\gamma(\omega)$ except that at $\omega = 55$ cm^{-1} this fit is a bit worse.

A second way to try to put less emphasis on the lowest datum point of HUR and to test their claim that at a given frequency the damping is proportional to the absolute temperature is discussed in this paragraph. The 491°C experimental data from $\omega = 20$ to 100 cm^{-1} and various extrapolations of the HUR data shown in Fig. 1 but multiplied by a number, Q, were used and a least-squares fit using the parameters ω_0 and Q was performed. This is a generalization of the above fits where the results in Fig. 1 are multiplied by $(491+273)/293 = 2.608$. Instead of Q, the results can be stated in terms of an effective temperature, T_e, which is the temperature that multiplies $\gamma(\omega)$ assuming[1] that it is proportional to the absolute temperature. We list T_e values in degrees centigrade for convenience. $T_e = +491$°C would mean that the least-squares fit requires $\gamma(\omega)$ to be proportional to absolute temperature. Table II shows the results of these fits and I comment only briefly: (1) is the two-parameter fit of the best fit from Table I. As can be seen, the MSD is about 4 times smaller. (2) is case a. (3) and (4) are two-parameter fits of the fits discussed in the above paragraphs where much less emphasis is attached to the lowest frequency point of experimental HUR data. Figure 3 shows the result from (4) which has the smallest MSD value. As can be seen, the fit is good on the low-frequency side of the data but poor on the high-frequency side where the damping is too small. This fit, (4), has a frequency-independent damping over the broadest range (20–50 cm^{-1}) with a value of 23.16 cm^{-1} which is of course close to the value found for γ assuming a frequency-independent damping. The other point to note is that T_e is close to room temperature in all these two-parameter fits.

TABLE II. Same as Table I but using two-parameter fits for the HUR data.

$\gamma(\omega)$	ω_0 (cm^{-1})	T_e (°C)	MSD
(1) Case b in Fig. 1	51.6	−13	145
(2) HUR but $\gamma = 42$ cm^{-1} for $20 \leq \omega \leq 45$ cm^{-1}	51.52	+12	126
(3) HUR but $\gamma = 32.8$ cm^{-1} for $20 \leq \omega \leq 45$ cm^{-1}	51.45	+22	106
(4) HUR but $\gamma = 21.5$ cm^{-1} for $20 \leq \omega \leq 50$ cm^{-1}	51.51	+43	95.5

There are two important, and independent, conclusions that one may draw from these results. Firstly, the fits using a frequency-independent damping constant are so good that there is very little room for any appreciable frequency dependence, at least using the model that leads to Eq. (1). Secondly, all of the extrapolations shown in Fig. 1, although quite reasonable in terms of the HUR data, give very poor fits to the experimental data, giving much too large a damping at low frequency. Or, as can be seen from Table II, the room-temperature damping is not, at a given frequency, proportional to temperature between 20 and 491°C.

I should like to point out that it is possible to resolve the problem discussed here. If between 20 and 491°C the frequency dependence of the HUR $\gamma(\omega, T = 20°\mathrm{C})$ result becomes considerably less frequency dependent, the result at 491°C could fit the experimental data considerably better. However, I hasten to add that the frequency-independent damping fits the data extremely well.

The discussion of fitting the right-angle Raman data with a damping constant, frequency independent or not, is complete. I use this last paragraph to clear up a misleading point in HUR associated with their Fig. 2. When one fits the right-angle Raman data with a frequency-independent damping constant, as done by BS, as large a range of ω is used as is possible. For example the range used by BS is 20–100 cm^{-1} and then extended[2b] to 9–100 cm^{-1}. The result is $\gamma_{\mathrm{BS}}(\omega, T$

$=491°C$), where ω refers to the *entire* frequency range. HUR have taken this value of damping constant and specialized it to $\gamma(\omega_0, T=491°C)$, i.e., only at a single frequency ω_0, then reduced it by $293/(491+273)$ to compare it to their polariton data, i.e., $\gamma(\omega=\omega_0, T=20°C)$. However, it makes no sense to single out an arbitrary frequency and specialize $\gamma_{BS}(\omega, T=491°C)$ to that frequency.

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[3]Throughout I have taken evenly spaced data points 5 cm^{-1} apart: either $\omega=20, 25, \ldots, 100\ cm^{-1}$ or for the truncated data $\omega=45, 50, \ldots, 100\ cm^{-1}$.

[4]This fit gives values for the intensity of 57.7 and 73.3 at $\omega=45$ and 55 cm^{-1}, respectively, while the data are 55 and 65 cm^{-1}, respectively, and the fit shown for $\gamma(\omega)$ in Fig. 2 gives values of 45.5 and 71.7.

Comment on Internal Field Distribution in Spin-Glasses

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It is shown that the low-temperature specific heat of spin-glasses in the random mean-field approximation is linear in the temperature, as observed experimentally, in disagreement with the conclusions of Held and Klein.

A recent Letter by Held and Klein[1] has reopened an old controversy[2] about the linear specific heat of spin-glasses at low temperatures. The argument under contention is that, since the probability distribution of internal fields $\vec{H}$ in spin-glasses $P(\vec{H})d^3H$ vanishes as $|\vec{H}|^2 d|\vec{H}|$ at low fields, the observed specific heat C which is linear in T at low temperatures is not explained by the mean-field approximation which yields $C \sim T^3$. [The classic mean-field derivation[3,4] of $C \sim T$ uses an Ising model which has finite $P(H=0)$]. The main aim of this Comment is to disagree with the conclusion, while accepting the premise. Indeed, a specific heat linear in temperature is obtained in the mean-field approximation, independent of the (Ising, Heisenberg) model.

The central results of Held and Klein's Letter, the probability distribution of internal fields at some spin site, $P(\vec{H})d^3H$ and $P(|\vec{H}|)d|\vec{H}|$ [Eqs. (16) and (18) of Ref. 1], have both been obtained and published previously[5,6] (see in particular Table I of Ref. 5), by an identical method. They are not disputed. The disagreement lies in the derivation of the specific heat. In Ref. 5, Adkins and the author have given a simple argument as to why Herring's conclusion is incorrect, and the specific heat is linear in T at low temperatures. Since the argument is short and has only appeared in somewhat obscure publications,[5,7] I shall repeat it here.

The magnetic energy of one particular spin in a random mean field $\vec{H}$ with probability distribution $P(\vec{H})d^3H$ is given by[3]

$$E_m = \int d^3H\, P(\vec{H})[\vec{m}(\vec{H}, T)\cdot\vec{H}], \tag{1}$$

where $\vec{m}(\vec{H}, T)$ is the magnetization of the spin under consideration,

$$\vec{m}(\vec{H}, T) = \hat{m} B(\hat{m}\cdot\vec{H}/T), \tag{2}$$

and B is the Brillouin function. Clearly $\vec{m}$ depends on the magnetic field only through the scalar product $\hat{m}\cdot\vec{H}$. It is thus possible to use cylindrical coordinates $d^3H = dH_z 2\pi H_\perp dH_\perp$ where the z direction is that of $\hat{m}$, however arbitrary in a spin-glass. Thus, with $\hat{m}\cdot\vec{H} = H_z$, Eq. (1) reads simply

$$E_m = \int dH_z H_z B(H_z/T)[2\pi \int dH_\perp H_\perp P(\vec{H})], \tag{3}$$

where $P(H_z) = 2\pi \int dH_\perp H_\perp P(\vec{H})$ is normalized: It is indeed the probability distribution of the component of the field along the direction $\hat{m}$ of the spin. Consequently, the relevant probability distribution for the magnetic energy and the specific heat $C = N(dE_m/dT)$ is not $P(|\vec{H}|)$ but $P(H_z)$, which is