Temperature dependence of acoustic attenuation in silicon

S. D. Lambade
Department of Physics, Y. C. College of Engineering, Wanadongri, Nagpur 441 110, India

G. G. Sahasrabudhe
Department of Physics, R. K. N. Engineering College, Nagpur 440 013, India

S. Rajagopalan
Department of Physics, Nagpur University, Nagpur 440 010, India
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This paper presents a comprehensive calculation of attenuation of ultrasonic waves in silicon. We have developed a computer program to calculate the attenuation using a much wider spectrum of thermal phonon modes than that used in Mason's pure-mode formulation. The program takes into consideration the dependence of Mason's integral on the cutoff frequency supported by the lattice. A comparison of these refined calculations with that using Mason's pure-mode scheme is presented for longitudinal waves along the [100], [110], and [111] directions and for transverse waves along the [100] and [110] directions in the temperature range 80–300 K. Use of experimentally measured relaxation times by Ilisavskii *et al.* in our program gives excellent agreement with experimental results. The results indicate essential validity of some of the objections raised against Mason's theory earlier by Barrett and Holland.

INTRODUCTION

Attenuation of high-frequency acoustic waves in solid crystals has been studied as a fundamental problem in solid-state physics with great interest in the last three decades.[1–6] The study of ultrasonic attenuation in semiconductors has also been reported extensively.[7–12] Mason[1] developed a theory to calculate the acoustic attenuation in solids with the help of second-order elastic constants (SOEC) and third-order elastic constants (TOEC). The theory has been used widely to account for the temperature variation of acoustic absorption in a variety of crystals.[10,12–15]

All these investigations employ Mason's scheme of calculation which is approximate in character as it considers interactions of the sound wave with only the pure thermal phonon modes (39 for longitudinal and 18 or 20 for shear waves) along principal directions for evaluating average Grüneisen number $\langle \gamma \rangle$ and the square average Grüneisen number $\langle \gamma^2 \rangle$. We have developed a computer program which takes into account interactions of the sound wave with a much wider spectrum of phonon modes for calculating these averages. Thus a more complete calculation using Mason's theory is possible with this program. The need for such a study has been stressed by several workers in the past.[12,16–21]

Mason's theory was critically examined by Barrett and Holland[3] shortly after it was put forward. However, it seems that these objections have not led to any serious modifications in calculation of ultrasonic attenuation based on Mason's theory. The present work is aimed at investigating the effect of inclusion of a more complete spectrum of the affected phonon modes and incorporation of the suggestions of Merkulov, Kovalenok, and Konovodehenko[18] in Mason's calculation of temperature dependence of attenuation in silicon for which the data on temperature dependence of TOEC is available.[22] We have estimated the attenuation in the temperature range 80–300 K using our program as well as using Mason's scheme for longitudinal waves along the [100], [110], and [111] directions and for transverse waves along the [100] and [110] directions. Comparison with the experimental variation of attenuation shows that these refinements in Mason's theory lead to a striking improvement in the quantitative agreement with the experimental attenuation for 9 out of the 11 cases studied.

THEORY

The attenuation of ultrasonic waves in semiconductors in the range $\omega\tau \ll 1$, where ω is the angular frequency of the acoustic wave and τ is the thermal relaxation time, is a result mainly of the interaction of acoustic phonons with thermal phonons. The two types of thermal attenuation in this region are (i) thermoelastic loss and (ii) Akhiezer loss.[23] Thermoelastic loss, which is a major source of attenuation in metals but contributes only about 4% in semiconductors, arises due to the flow of thermal energy from the compressed hotter part of the expanded cooler part associated with the compressional wave. This flow causes an attenuation given by

$$A\,(Np/\mathrm{cm}) = [\omega^2 \langle \gamma \rangle^2 KT]/[2\rho v_l^5]\ , \qquad (1)$$

where $\langle \gamma \rangle$ is the average Grüneisen constant, K is the thermal conductivity, T is the absolute temperature, ρ is the density, and v_l is the longitudinal velocity. Akhiezer loss originates due to the sudden application of strain causing phonons propagating in different directions to have different temperatures. These changes in temperatures result in a thermal energy storage proportional to the square of the applied strain and hence to an equivalent increase in the elastic modulii associated with the strain. Akhiezer's theory was modified by Mason and Bateman who proposed a scheme of evaluation involving the use of TOEC to determine the energy stored by the phonon-mode temperature separations, together with a relaxation time τ to equilibrate this energy. Their expression for attenuation is

$$A\,(Np/\mathrm{cm}) = [E_0 D/6\rho v^3][\omega^2\tau/(1+\omega^2\tau^2)] , \tag{2}$$

where

$$D = 3\left[3\sum_i (\gamma_i^j)^2/n - \langle\gamma\rangle^2 CT/E_0\right] , \tag{3}$$

is the nonlinearity constant. C is the specific heat per unit volume, v is the appropriate wave velocity, and E_0 is the thermal energy content per unit volume. In the high-temperature approximation $E_0 = nE_i$, where n is the total number of modes and E_i is the thermal energy of the modes of type i given by

$$E_i = [3hN_i/\nu_{qi}^3]\int_0^{\nu_{qi}} \{\nu^3 d\nu/[\exp(h\nu/kT)-1]\} , \tag{4}$$

where N_i is the total number of modes in the sector of type i and ν_{qi} is the limiting or cutoff frequency supported by the lattice. Mason considered the nonlinearity constant independent of temperature as the experimental data on temperature dependence of TOEC was not available at that time. Recently Breazeale and Philip[22] presented the experimental determination of temperature variation of all six TOEC for Si and Ge using the simple harmonic generation technique and Keating model.[24] The temperature dependence of D has been studied using this data.[25–27]

The relaxation times τ_l and τ_s for longitudinal and shear waves, respectively, are given by

$$\tau_l = 6K/(C\langle v\rangle^2) \tag{5}$$

and

$$\tau_s = \tau_l/2 . \tag{6}$$

The Debye average velocity $\langle v\rangle$ is given by

$$\langle v\rangle^{-3} = [2v_s^{-3} + v_l^{-3}]/3 , \tag{7}$$

where v_s is the shear-wave velocity.

The mode Grüneisen numbers γ_i^j can be expressed[28] in terms of SOEC and TOEC as

$$\gamma_i^{jk} = (1/2W)[2WU_jU_k + (C_{jkmn} + C_{jkmunv}U_uU_v)N_mN_n] \tag{8}$$

with

$$W = C_{munv}N_mN_nU_uU_v , \tag{9}$$

where N's and U's are the direction cosines of the propagation direction and polarization direction, respectively, in the acoustic mode i and C's are the elastic constants.

Grüneisen numbers along an axis other than cube axes are obtained by

$$\gamma_i^{j'k'} = [\partial_{j'}/\partial x_j][\partial x_{k'}/\partial x_k]\gamma_i^{jk} , \tag{10}$$

where $\partial x_{j'}/\partial x_j$ are the direction cosines between the new axes and the old axes.

Mason considers the actuation of only the phonon modes along the crystallographic directions associated with the propagation of sound wave for calculating the averages $\langle\gamma\rangle$ and $\langle\gamma^2\rangle$. The number of modes considered is thus only 39 in case of longitudinal waves and is 18 or 20 in case of shear waves. This is only an approximation for the complete integral for all directions which requires the use of a computer and a complicated program.

Shortly after Mason's theory was put forward Barrett and Holland[3] pointed out difficulties, both conceptual and in formulation, in the theory. One of the major contentions of Mason which was objected to was that the integral

$$I(\nu_{qi}) = \int_0^{\nu_{qi}} \{\nu^3 d\nu/[\exp(h\nu/kT)-1]\} \tag{11}$$

in Eq. (4) was taken to be essentially independent of ν_{qi} in deriving Eq. (2). In our computational scheme it is possible to remove this difficulty by explicitly estimating the effect of dependence of the integral on ν_{qi}. This was pointed out by Merkulov[18] and leads to a correction factor

$$M_i = 1 - [\nu_{qi}/3I(\nu_{qi})][\partial I(\nu_{qi})/\partial\nu_{qi}] .$$

This factor is easily derived. The ν_{qi} derivative of the integral in Eq. (11) above is just the integrand evaluated at ν_{qi}, i.e.,

$$\partial I(\nu_{qi})/\partial\nu_{qi} = \nu_{qi}^3/[\exp(h\nu_{qi}/kT)-1] .$$

This leads to

$$M_i = 1 - 1/\{3I'(\nu_{qi})[\exp(h\nu_{qi}/kT)-1]\} ,$$

where the integral $I'(\nu_{qi})$ is given by

$$I'(\nu_{qi}) = \int_0^1 d\mu\,\mu^3/[\exp(h\nu_{qi}\mu/kT)-1] ,$$

TABLE I. Temperature dependence of various parameters used in the calculation of attenuation.

T (K)	α (10^{-6}/K)	θ_D/T	C (10^7 erg/cm^{3}K)	E_0 (10^7 erg/cm^3)
80	−0.5012	8.0857	0.2786	5.8817
100	−0.3000	6.4686	0.4711	13.3380
120	−0.1806	5.3903	0.6728	24.7910
140	0.3257	4.6197	0.8620	40.1708
160	0.6805	4.0418	1.0293	59.1335
180	1.0479	3.5917	1.1725	81.2163
200	1.4000	3.2318	1.2932	105.9377
220	1.7131	2.9373	1.3945	132.8998
240	1.9821	2.6917	1.4789	161.6593
260	2.2069	2.4841	1.5499	192.0210
280	2.3934	2.3060	1.6096	223.6593
300	2.5540	2.1516	1.6601	256.3640

TABLE II. Temperature dependence of velocity and relaxation time for different directions. For the [110] direction v_{s1} denotes the velocity for the wave polarized along the [1$\bar{1}$0] direction. Velocity v_{s2} for the [110] shear wave polarized in the [001] direction is the same as v_s [100]. For the [100] shear wave $\tau_s = \tau_l/2$.

T	Velocity (10^5 cm/sec)					Relaxation time (10^{-10} sec)				
	[100]		[110]		[111]	[100]	[110]			[111]
(K)	v_l	v_s	v_l	v_{s1}	v_l	τ_l	τ_l	τ_{s1}	τ_{s2}	τ_l
80	8.4725	5.8586	9.1728	4.6868	9.3947	7.369	4.951	3.263	2.229	10.916
100	8.4708	5.8582	9.1710	4.6867	9.3928	6.000	3.437	2.229	1.587	4.416
120	8.4687	5.8576	9.1690	4.6861	9.3908	5.167	2.616	1.691	1.208	2.137
140	8.4657	5.8564	9.1660	4.6851	9.3879	4.450	2.134	1.399	1.000	1.211
160	8.4623	5.8553	9.1626	4.6843	9.3844	4.000	1.823	1.134	0.833	0.802
180	8.4582	5.8533	9.1584	4.6826	9.3802	3.632	1.459	0.972	0.727	0.583
200	8.4540	5.8515	9.1540	4.6812	9.3758	3.381	1.234	0.833	0.600	0.453
220	8.4493	5.8497	9.1495	4.6795	9.3712	2.981	1.111	0.744	0.542	0.367
240	8.4443	5.8474	9.1443	4.6776	9.3661	2.775	0.978	0.629	0.470	0.307
260	8.4400	5.8458	9.1400	4.6762	9.3617	2.530	0.855	0.560	0.425	0.265
280	8.4350	5.8437	9.1350	4.6745	9.3567	2.366	0.761	0.500	0.400	0.234
300	8.4297	5.8415	9.1299	4.6720	9.3516	2.250	0.673	0.443	0.345	0.209

this factor can be incorporated into a numerical integration scheme to find the required averages of Grüneisen numbers.

The computer program developed by us for this purpose is based on Brugger-Fritz[29] scheme of integration over the length of the wave vector followed by double angular integration over all directions. We have checked the consistency of results using different single and multiple integration routines. The program calculates the averages of first and second power of mode Grüneisen numbers required by Mason's procedure. The program is flexible in the sense that weighted averages required for other schemes like those of Maris[2] and Merkulov, Kovalenok, and Konovodehenko[18] can be calculated by making appropriate modifications. The program also incorporates the Merkulov correction factor mentioned above.

RESULTS AND DISCUSSION

In our calculation we have taken into account the temperature dependence of each parameter in expressions (1), (2), and (3) in evaluating the temperature variation of attenuation. Temperature dependence of density was calculated using the room-temperature density, $\rho = 2.331$ gm/cm^3 and the data on the temperature-dependent thermal-expansion coefficient (α).[30] The wave velocities were calculated using the relevant expressions[31] and the temperature dependence of SOEC.[32] Debye velocity and relaxation time along the [111] direction were computed using Eqs. (5)–(7) and the temperature-dependent thermal conductivity data.[33] For other directions the relaxation times used are those reported by Ilisavskii and Sternin[34] from the identification of the kink in the frequency-attenuation plots at various temperatures. For [100] shear waves we have used relaxation times which

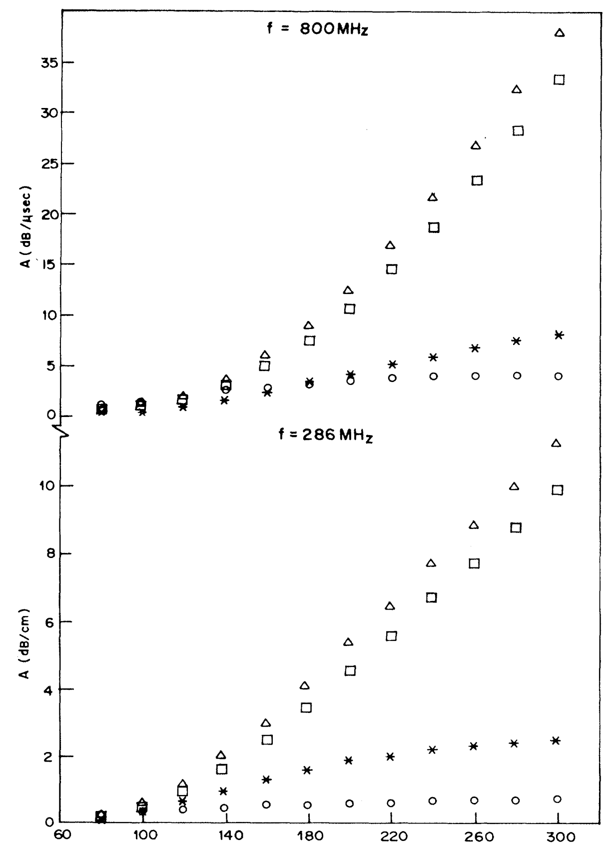

FIG. 1. Temperature dependence of ultrasonic attenuation for longitudinal waves along the [100] direction at frequencies $f = 800$ MHz and $f = 286$ MHz. △: values calculated using Mason's scheme, * and □: values calculated using our computer program with and without Merkulov's correction factor, respectively, ○: Experimental values from Ref. 8 for 800 MHz and Ref. 7 for 286 MHz.

TABLE III. Values of $\langle \gamma \rangle$ and $\langle \gamma^2 \rangle$ at room temperature for different directions.

Direction of propagation	Direction of polarization	$\langle \gamma \rangle$ a	$\langle \gamma \rangle$ b	$\langle \gamma \rangle$ c	$\langle \gamma^2 \rangle$ a	$\langle \gamma^2 \rangle$ b	$\langle \gamma^2 \rangle$ c
100	100	0.6368	0.6531	0.1315	1.0164	0.9406	0.1795
100	001				0.0601	0.0732	0.0146
110	110	0.6812	0.6531	0.1315	0.8382	0.7255	0.1533
110	$1\bar{1}0$				0.9338	0.9320	0.1196
110	001				0.0783	0.0732	0.0148
111	111	0.7252	0.6531	0.1315	0.7848	0.6540	0.1445

[a]Calculation using Mason's pure-mode scheme.
[b]Calculation using our computer program without Merkulov's correction factor.
[c]Calculation using our program with Merkulov's correction factor.

are half the relaxation times for [100] longitudinal waves. The Debye characteristic temperatures, θ_D were calculated at different temperatures using de Launay's technique[35] which were further used to estimate the temperature variation of energy and specific heat.[30] The temperature dependence of various parameters used in the calculation of attenuation is shown in Tables I and II.

For estimation of attenuation using Mason's pure-mode scheme we have used expressions for γ_i^j tabulated by Mason.[7,12,16] For longitudinal waves along the [111] direction we use refined expressions for γ_i^j deduced by Merkulov, Kovalenok, and Konovodehenko.[18] For calculation of attenuation using our program the number of directions for which γ_i^j are evaluated is determined by a convergence criterion for the numerical approximation of the full integral. The minimum number of directions considered is 800. For shear waves $\langle \gamma_i^j \rangle$ was found to be consistently less than 10^{-6} with the imposed convergence

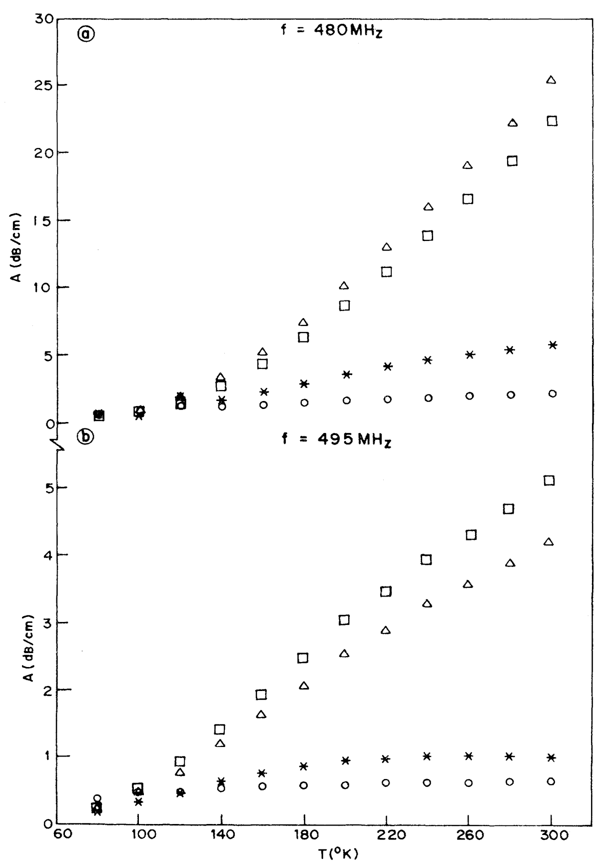

FIG. 2. Same as in Fig. 1 for (a) longitudinal and (b) shear waves along the [100] direction for $f=480$ and 495 MHz respectively. Experimental values from Ref. 7 for both cases.

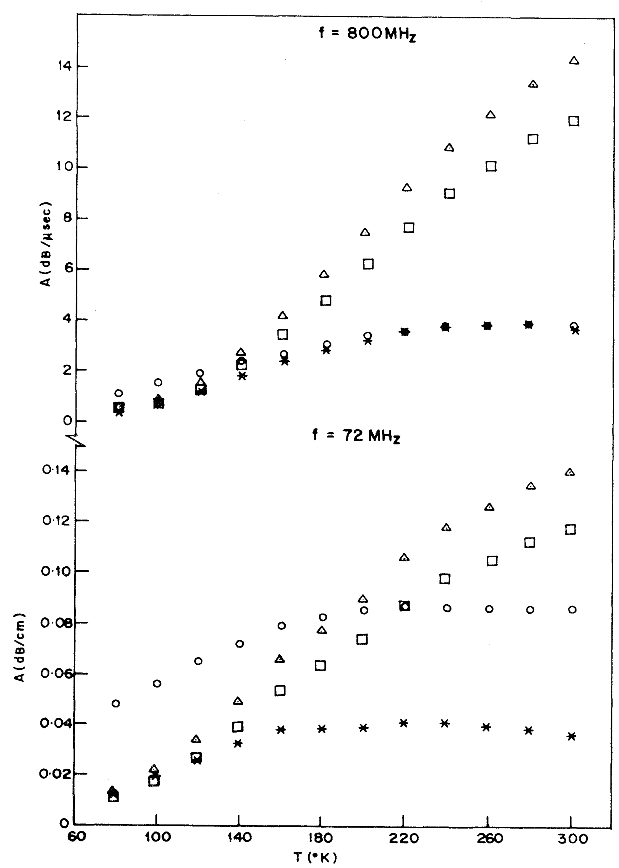

FIG. 3. Same as in Fig. 1 for longitudinal waves along the [110] direction for $f=800$ and 72 MHz. Experimental values from Ref. 8 for 800 MHz and Ref. 36 for $f=72$ MHz.

criterion. This points to the essential correctness of the numerical approximation to the full integral. Table III illustrates the comparison of $\langle \gamma \rangle$ and $\langle \gamma^2 \rangle$ for different directions at room temperature.

The attenuation calculated using both these approaches for different directions are shown in Figs. 1–6. The attenuation of longitudinal waves also includes the contribution of thermoelastic loss calculated using Eq. (1). There is no thermoelastic loss for shear waves. For waves propagating along [110] direction Mason[16] has suggested that γ_i^j should be weighted by appropriate relaxation time. Merkulov has extended this refinement in relative weighting of γ_i^j to waves along [111] direction. The correct weighting of γ_i^j by different relaxation times is taken into account in our program also for evaluation of attenuation for these directions.

From the results presented in Figs. 1–6 it is clear that the calculation based on the correction factors resulting from the dependence of integral (11) on the cutoff frequency supported by lattice and the use of the collective phonon relaxation time obtained from the condition $\omega\tau = 1$ leads to quite a close agreement with the experimental attenuation, except for [110] shear waves polar-

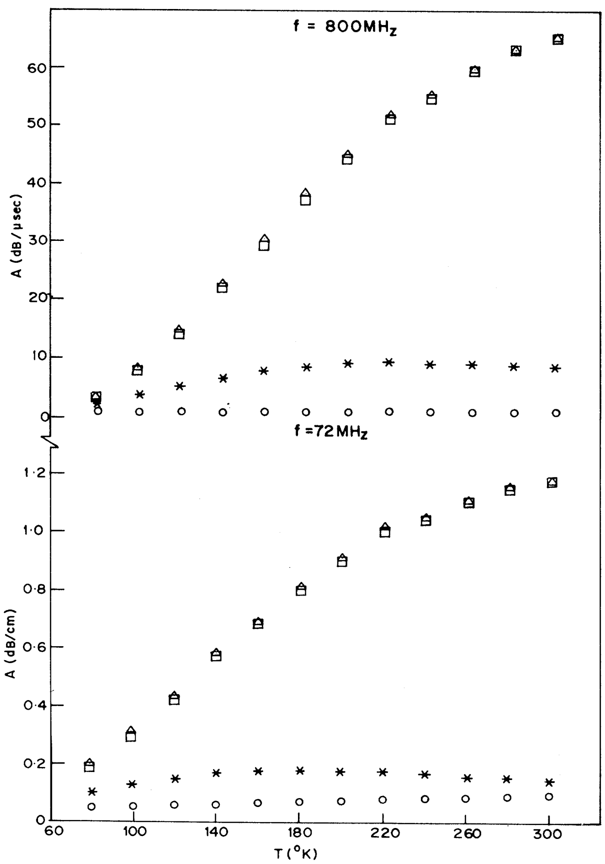

FIG. 4. Same as for Fig. 1 for shear waves along the [110] direction polarized along the [1$\bar{1}$0] direction for $f = 800$ and 72 MHz. Experimental values from Ref. 8 for 800 MHz and Ref. 36 for 72 MHz.

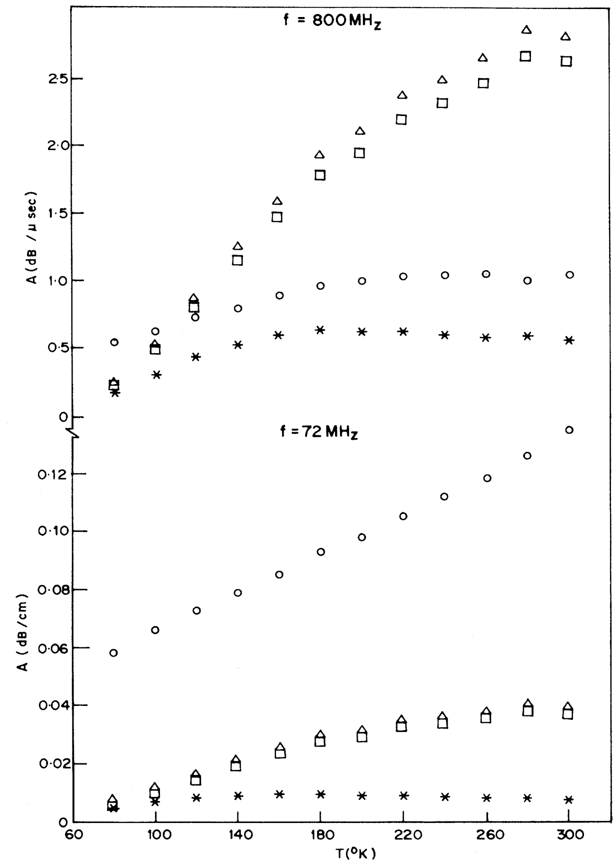

FIG. 5. Same as for Fig. 1 for shear waves along the [110] direction polarized along the [001] direction for $f = 800$ and 72 MHz. Experimental values from Ref. 8 for 800 MHz and Ref. 36 for 72 MHz.

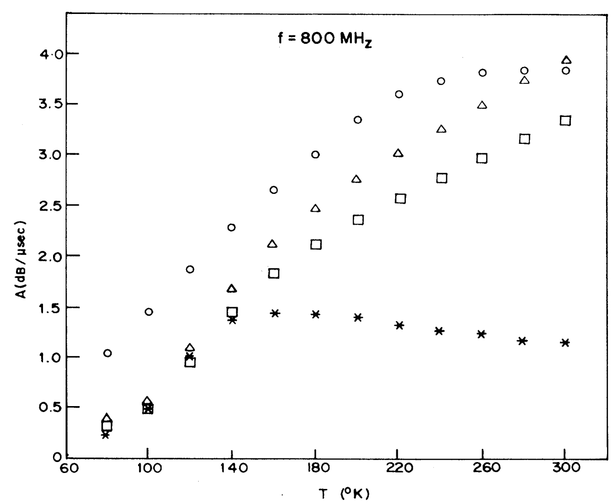

FIG. 6. Same as for Fig. 1 for longitudinal waves along [111] direction for $f = 800$ MHz. Experimental values from Ref. 8.

ized along the [001] direction for 72 MHz and for longitudinal waves along the [111] direction. For longitudinal waves along the [111] direction, better agreement can be expected if the data on experimental relaxation times is available. Here it may be recalled that apart from the ν_{qi} dependence of integral (11) Barrett and Holland have pointed out other factors which are important for Si. These include dependence of γ's on the wave vector as well as polarization.

As our program facilitates calculation of attenuation using other rigorous theories outlined by Maris[2] with only little modification, a more complete theoretical investigation of attenuation in various solids using different models is being undertaken. The experiments on the frequency dependence of attenuation at various temperatures, which can be used to find the collective phonon relaxation times in other solids, would be quite useful for these investigations.

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