

small r , $H_I(m_2^*)$ goes to zero, the self-energy of the vacancy being canceled, while as $r \rightarrow \infty$ the only energy is the vacancy-lattice self-energy.

To continue in the spirit of Haken we fit $H_I(m_2^*)$ to an exponential such that it behaves properly for large and small r , the result being

$$H_I(m_2^*) \approx (e^2/2\epsilon^*r)[1 - e^{-2r/a}] - (e^2/\epsilon^*a). \quad (A7)$$

The exponential fit is not too good for $r \lesssim a$ but in the region of interest is sufficiently accurate.

The last terms in Eqs. (A4) and (A7) are the self-energies of the electron and vacancy due to the polariza-

tion of the lattice. These negative terms outside the potential well are equivalent to a change in the zero of energy inside, and in fact these terms derived from polaron theory are analogous to the last term in W , Eq. (5). Thus to be perfectly consistent one should, when using the Haken dielectric constant, replace the last term in W by the quantity $(e^2/\epsilon^*)[(v/2) + (1/a)]$. This we have not done. For the large-orbit state the terms in question are approximately equal, since $v/2 \ll 1/a$ and $e^2/\epsilon^*a \approx e^2/\epsilon^*R$. However, in treating a smaller-radius excited state one should use the Haken self-energy.

Coupled-Optical-Phonon-Mode Theory of the Infrared Dispersion in BaTiO₃, SrTiO₃, and KTaO₃*

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The independent-oscillator model fails to predict the dielectric behavior of the high-dielectric-constant materials BaTiO₃, SrTiO₃, and KTaO₃ in certain infrared-frequency regions near the three infrared-active modes. A more general classical model is proposed with mode coupling. The model has one additional parameter for each pair of modes that are coupled, and gives decreased (or increased) dielectric loss in certain regions between the modes, compared with the independent oscillator model. Very satisfactory fits to reflectivity data for the above materials are obtained using the coupled-mode theory. In a mechanical analog of the model, the coupling element may be either a spring or a dashpot, since the two cases are shown to be equivalent. For the above materials, however, there is a physically interesting simplification in the spring-coupling form, since two of the otherwise arbitrary parameters are zero. The result suggests that the damping is best viewed as applying to the total polarization rather than to the individual normal modes.

INTRODUCTION

IN an insulating crystal with more than one infrared active mode, the reflectivity in the *reststrahlen* region is often quite well reproduced by a formula involving the sum of contributions from independent classical oscillators.¹ There are notable exceptions, however. The high dielectric constant materials BaTiO₃, SrTiO₃, and KTaO₃ provide a striking example. In fitting independent classical oscillator expressions one is immediately faced with compromises when choosing the damping constants for some modes. For example, near 21- μ wavelength in the case of SrTiO₃, the reflectivity shows a dip to a value of less than 1%. The low damping indicated by this dip is inconsistent with the large damping nearby.² Figure 1 shows SrTiO₃ reflectiv-

ity data and the best over-all classical oscillator fit.³ The fit can be improved near 21 μ by decreasing the damping constant of the highest frequency mode. This effect is shown in the figure—there is an improved fit near the minimum but a poorer fit elsewhere. The fit can also be improved by decreasing the damping constant of the lowest frequency mode. Again there is improvement only over a small wavelength interval. Similarly, the rather square reflectivity shoulder extending from 22 to 26 μ cannot be reproduced at all by the classical oscillator formula without choices of damping constants which spoil the fit elsewhere.

There are two separate difficulties involved in attempting to go beyond the classical independent oscillator model. One problem is to correctly describe

on the high-frequency side of a mode. If ϵ'' , the imaginary part of the dielectric constant, is zero here, the reflectivity will dip to zero. The dip then provides a sensitive measure of ϵ'' which, in turn, depends directly on γ , the damping constant.

³W. G. Spitzer, R. C. Miller, D. A. Kleinman, and L. E. Howarth, Phys. Rev. **126**, 1710 (1962).

* Work supported in part by the National Science Foundation.

¹W. G. Spitzer and D. A. Kleinman, Phys. Rev. **121**, 1324 (1961).

²In general, such dips occur at the frequency where ϵ' , the real part of the dielectric constant, is passing through the value $\epsilon' = 1$

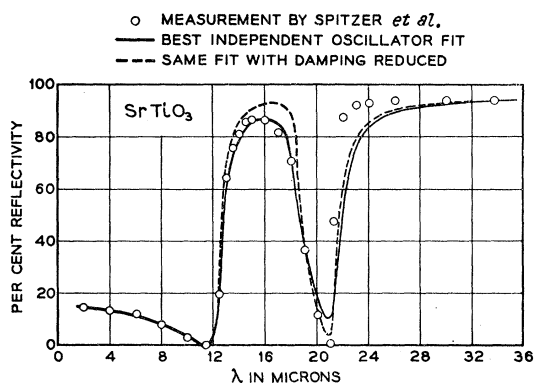


FIG. 1. Reflectivity of SrTiO₃. The curves were calculated using three independent classical oscillators. For the dashed curve the damping of the oscillator at 18.4 μ was reduced approximately a factor of 3 to improve the fit near the reflectivity minimum.

the frequency dependences of the damping "constant." The calculation of this frequency dependence^{4,5} involves the knowledge of the phonon spectrum of the crystal and the nature of the anharmonic interactions, and is beyond the scope of the present paper.

The second difficulty concerns the independence of modes. In crystals having more than one $\mathbf{k}=0$ transverse optical mode which interacts with light, a quantum mechanical calculation shows that the damping of the different modes is not necessarily independent. The interaction of the damping of different modes can be represented by a classical model. This classical model is developed for the case of two $\mathbf{k}=0$ transverse phonon-optical modes in the present paper. The use of this model to fit the reflectivities of BaTiO₃, SrTiO₃, and KTaO₃ produces a major improvement in the agreement between experiment and theory. In addition, it provides insight into the relation between the normal modes and the damping process.

Interaction Damping—The Quantum Mechanical Cause

The usual damping of optical modes is due to the process

optical phonon \rightarrow several other phonons,

induced by anharmonic terms in the lattice potential-energy expansion. Similarly, optical absorption associated with an optical-phonon transition is due to the process

photon $l \rightarrow$ optical phonon $i \rightarrow$ several other phonons f
(and similar processes involving other orders of events).

⁴ M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Clarendon Press, Oxford, 1954), Sec. 46.

⁵ V. S. Vinogradov, *Fiz. Tverd. Tela* **3**, 1726 (1961) [English transl.: *Soviet Phys.—Solid State* **3**, 1249 (1961)]; R. F. Wallis and A. A. Maradudin, *Phys. Rev.* **125**, 1277 (1962); M. Lax, *Phys. Chem. Solids* **25**, 487 (1964).

If H_{li} is the matrix element for converting a photon into an optical phonon of type i , and H_{if} is the matrix element for converting an optical phonon i into the final state f , then the transition amplitude for creating several other phonons f via intermediate phonon i is given by

$$H_{li}H_{if}/(E_l - E_i),$$

where E_i is the energy of an optical phonon of type i , and E_l is the energy of the photon. This expression would have to be modified slightly for E_l very near E_i . The net transition probability for getting from a photon to state f is, of course, proportional to the square of the absolute value of the sum of the transition amplitudes via all intermediate states i ; that is, to

$$|\sum_i H_{li}H_{if}/(E_l - E_i)|^2. \quad (1)$$

If the contributions of different intermediate state phonons i were independent, this sum would necessarily reduce to

$$\sum_i |H_{li}H_{if}/(E_l - E_i)|^2. \quad (2)$$

Under certain special cases, this reduction occurs; for example, if one term completely dominates the sum for a given E_l , or if H_{if} couples one state i to states f , and other states i' only to f' , i'' to f'' , etc. In general, however, (1) not (2) is the appropriate expression.^{6,7} In this general case, the optical absorption due to the wings of different phonon lines are not additive. It is to be expected that such interference will manifest itself in the vicinity of $E_l \approx E_i$, although simple formulas like (1) are then not sufficient.

Interaction Damping—The Classical Approach

The fundamental reason that a simple classical one-level resonance expression for the dielectric constant performs as well as it does for a crystal having but one $\mathbf{k}=0$ optical branch is that the region of primary importance is relatively narrow in frequency. The simple classical equation of motion

$$\ddot{y}_1 + \Gamma_1 \dot{y}_1 + k_1 y_1 = e_1 E, \quad (3)$$

with $P = e_1 y_1$, is the most general one mode equation possible for the polarization P as a function of electric field E , if by "one mode" we mean a dielectric response function having only one pair of simple poles. For positive Γ_1 , the system is of course dissipative.

Let us define a two-mode dielectric response function as a response function having two pairs of poles. The most general such dielectric response function which can be generated from a set of equations of motion of two variables (consistent with all motions of the variables

⁶ J. J. Hopfield, *Phys. Chem. Solids* **22**, 63 (1961).

⁷ G. Baym and V. Ambegaokar, *Phys. Chem. Solids* (to be published).

being dissipative) can be written⁸

$$\begin{aligned} \ddot{y}_1 + \Gamma_1 \dot{y}_1 + (k_1 + k_{12})y_1 - k_{12}y_2 &= e_1 E, \\ \ddot{y}_2 + \Gamma_2 \dot{y}_2 + (k_2 + k_{12})y_2 - k_{12}y_1 &= e_2 E, \\ P &= y_1 e_1 + y_2 e_2. \end{aligned} \quad (4)$$

Here, y_1 and y_2 are the displacements of particles 1 and 2. The Γ and k coefficients represent damping and restoring forces and the e coefficients, effective charge. The only restrictions on the real constants Γ_1 , Γ_2 , k_1 , k_2 , k_{12} , e_1 , e_2 are that Γ_1 , Γ_2 , $(k_1 + k_2 + 2k_{12})$, and $[k_1 k_2 + k_{12}(k_1 + k_2)]$ be positive. The usual dielectric model for two independent oscillators is identical to the above model with $k_{12} = 0$.

The above equations of motion can be written in the following equivalent way:

$$\begin{aligned} \ddot{x}_1 + (\gamma_1 + \gamma_{12})\dot{x}_1 - \gamma_{12}\dot{x}_2 + \omega_1^2 x_1 &= z_1 E, \\ \ddot{x}_2 + (\gamma_2 + \gamma_{12})\dot{x}_2 - \gamma_{12}\dot{x}_1 + \omega_2^2 x_2 &= z_2 E, \\ P &= z_1 x_1 + z_2 x_2, \end{aligned} \quad (5)$$

with the equivalence given by

$$\begin{aligned} u &\equiv \begin{pmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{pmatrix}, \\ \begin{pmatrix} x_1 \\ x_2 \end{pmatrix} &= u \begin{pmatrix} y_1 \\ y_2 \end{pmatrix}, \quad \begin{pmatrix} z_1 \\ z_2 \end{pmatrix} = u \begin{pmatrix} e_1 \\ e_2 \end{pmatrix}, \\ \begin{pmatrix} \gamma_1 + \gamma_{12} & \gamma_{12} \\ y_{12} & \gamma_2 + \gamma_{12} \end{pmatrix} &= u \begin{pmatrix} \Gamma_1 & 0 \\ 0 & \Gamma_2 \end{pmatrix} u^{-1}, \\ \begin{pmatrix} \omega_1^2 & 0 \\ 0 & \omega_2^2 \end{pmatrix} &= u \begin{pmatrix} k_1 + k_{12} & k_{12} \\ k_{12} & k_2 + k_{12} \end{pmatrix} u^{-1}. \end{aligned} \quad (6)$$

Such a unitary transformation which diagonalizes the

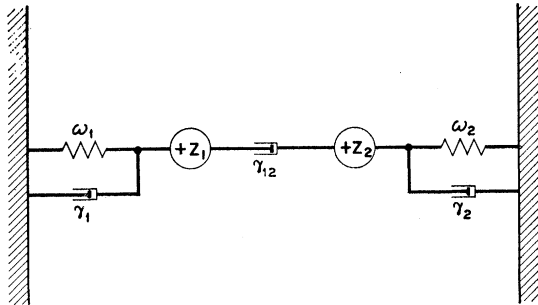


FIG. 2. Mechanical model of two optic mode oscillators with interaction damping. The dashpots γ_1 , γ_2 , and γ_{12} provide damping forces proportional to velocity.

⁸ To simplify notation we have included the reduced mass (m) of the mode in the definition of the mode amplitudes, force constants, and charges. In these and following equations force constants have dimensions of (frequency)²; damping factors and effective charges have dimensions of (frequency). The effective charges here are equal to $e^*/(mV)^{1/2}$, where e^* is the usual effective charge defined when the local electric field is taken to be the macroscopic field and V is the volume of the unit cell.

force constant matrix can always be found. θ is given by

$$\cot^2\theta + [(k_2 - k_1)/k_{12}]\cot\theta - 1 = 0.$$

The mechanical model corresponding to Eq. (5) is shown in Fig. 2. We can quickly see the importance of such a model in the present work. For a driving field of nearly any frequency, the three dashpots γ_1 , γ_2 , and γ_{12} shown in Fig. 2 contribute to the losses. If the two particles are oppositely charged there will be some frequency intermediate to the two resonant frequencies, however, where the particles move together causing the γ_{12} dashpot to be inactive. At this frequency the losses will be reduced. This is the situation we anticipate near 21μ in SrTiO₃. The model originally was conceived in this latter form [Eq. (5)] where the mode coupling is a dashpot, and it has been found much easier to fit data with this form. Equation (4) with only harmonic-force coupling was found useful in checking the over-all dissipative nature of the solution when the negative γ 's were encountered. Of course, an arbitrary number of equivalent intermediate forms could be written involving both kinds of coupling by using unitary transformations u which diagonalize neither the force-constant matrix nor the damping matrix. We return to the significance of the transformation between the equivalent forms after obtaining the explicit solutions to Eq. (5).

We shall search for the harmonic solutions

$$E, x_1, x_2 \sim e^{i\omega t}.$$

Inserting these dependences into Eq. (5) and solving, we obtain

$$\begin{aligned} x_1 &= \frac{z_1 E + \frac{i\omega\gamma_{12}z_2 E}{\omega_2^2 - \omega^2 + i\omega(\gamma_2 + \gamma_{12})}}{\omega_1^2 - \omega^2 + i\omega(\gamma_1 + \gamma_{12}) + \frac{\omega^2\gamma_{12}^2}{\omega_2^2 - \omega^2 + i\omega(\gamma_2 + \gamma_{12})}} e^{i\omega t}, \\ x_2 &= \left(\begin{array}{l} 1 \rightarrow 2 \\ \text{Same with} \\ 2 \rightarrow 1 \end{array} \right). \end{aligned}$$

We compute the dielectric constant by adding the currents arising from the driven motion of each oscillator plus the usual background term ϵ_∞ arising from any much higher frequency modes such as excitons, and from the vacuum.

$$\epsilon = \epsilon_\infty + 4\pi P/E = \epsilon_\infty + \epsilon_1 + \epsilon_2,$$

where

$$\epsilon_1 = 4\pi z_1 x_1/E, \quad \epsilon_2 = 4\pi z_2 x_2/E.$$

Substituting, we obtain

$$\epsilon_1 = \frac{4\pi z_1^2 + i4\pi z_1 z_2 \omega \gamma_{12} / [\omega_2^2 - \omega^2 + i\omega(\gamma_2 + \gamma_{12})]}{\omega_1^2 - \omega^2 + i\omega(\gamma_1 + \gamma_{12}) + \omega^2 \gamma_{12}^2 / [\omega_2^2 - \omega^2 + i\omega(\gamma_2 + \gamma_{12})]} \quad (7)$$

for the dielectric constant. Interchange of subscripts gives ϵ_2 .

DISCUSSION

Examining Eq. (7), we note that when $\gamma_{12}=0$ we have the usual noninteracting classical oscillator. If $\gamma_{12}\neq 0$ and $z_2=0$ (an infrared active mode interacting with an infrared inactive mode) there can still be interaction effects on ϵ_1 via the last term in the denominator. A third point is that the sign of the second term in the numerator depends on the signs of the effective charges of the two interacting modes. The independent mode formula is insensitive to the sign of the effective charges since only z_1^2 and z_2^2 appear. We thus have a new method of analyzing ion motions for comparison with a theoretical normal mode analysis if fits are carried out using the interacting-mode theory. Both possibilities for the net sign of $z_1 z_2$ are encountered in the fits described below.

Figures 3, 4, 5, and 6 show the fits obtained for SrTiO₃, BaTiO₃, and KTaO₃ using interaction damping. There is a great improvement in the region that could not be fitted by any choice of parameters using uncoupled oscillators. In computing the curves only two modes were taken to interact because of the very cumbersome equations for the 3 interacting mode case. A third classical mode was added in each case, however, as these perovskite structure materials all

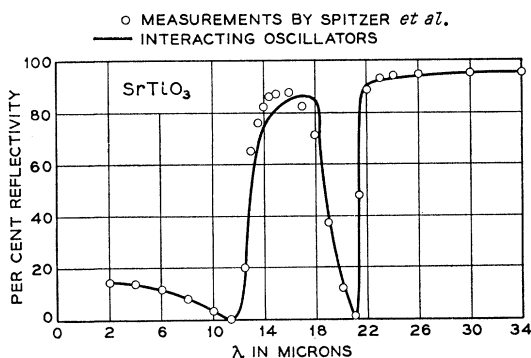


FIG. 3. Interaction model fit (solid curve) to SrTiO₃ reflectivity data.

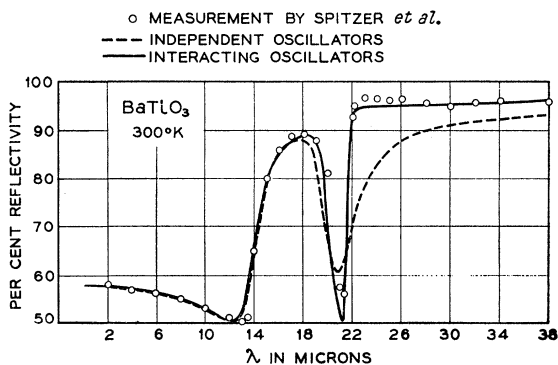


FIG. 4. Independent mode (dashed curve) and interacting mode fits to BaTiO₃.

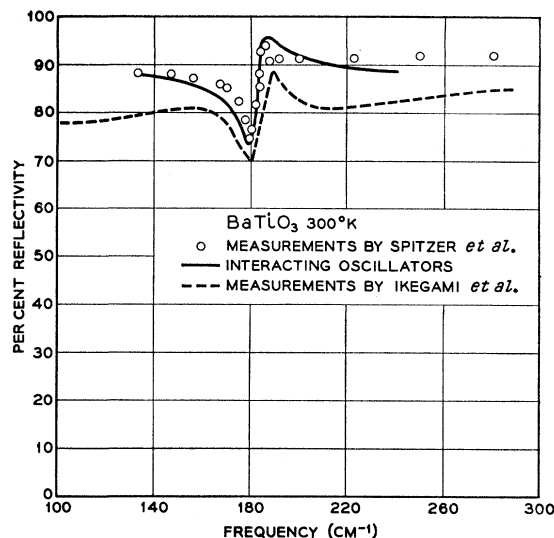


FIG. 5. Interaction mode fit to BaTiO₃ reflectivity near 180 cm⁻¹.

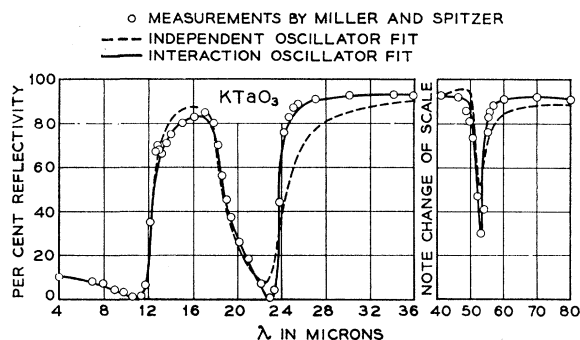


FIG. 6. Independent mode (dashed curve) and interacting mode fits to KTaO₃.

have three optically active modes. To compute the reflectivity R we use $\epsilon = \epsilon_1 + \epsilon_2 + \epsilon_3 + \epsilon_\infty$, where ϵ_1 and ϵ_2 are given by Eq. (7),

$$\epsilon_3 = 4\pi z_3^2 / (\omega_3^2 - \omega^2 + i\omega\gamma_3) \text{ (independent mode),}$$

and

$$R = |(1 - \sqrt{\epsilon}) / (1 + \sqrt{\epsilon})|^2.$$

Table I gives the mode parameters used in each figure. SrTiO₃ does not have an anomalous reflectivity near 178 cm⁻¹ so there seems no need to invoke the additional coupling of the two lowest frequency modes to explain the data. In KTaO₃, however, there is considerable deviation from independent oscillator behavior at low frequencies. The two lowest frequency modes have been coupled to produce the excellent fit shown on the right-hand side of Fig. 6. In BaTiO₃ (Fig. 5) the departure at low frequencies from independent mode behavior is small, though Ikegami *et al.* do show a larger effect in

TABLE I. Oscillator parameters used in calculations.

Fig. No.	Material	Interacting modes ^a						Third independent mode			ϵ_∞		
		$\frac{4\pi z_1^2}{\omega_1^2}$	$\frac{\omega_1}{(\text{cm}^{-1})}$	$\frac{\gamma_1 + \gamma_{12}}{\omega_1}$	$\frac{4\pi z_2^2}{\omega_2^2}$	$\frac{\omega_2}{(\text{cm}^{-1})}$	$\frac{\gamma_2 + \gamma_{12}}{\omega_2}$	$\frac{\gamma_{12}}{(\omega_1 \omega_2)^{1/2}}$	$\frac{4\pi z_3^2}{\omega_3^2}$	$\frac{\omega_3}{(\text{cm}^{-1})}$		$\frac{\gamma_3}{\omega_3}$	
1	solid	SrTiO ₃	311	88	0.4	1.56	544	0.05	0	3.6	178	0.04	5.2
1	dashed	SrTiO ₃	311	88	0.4	1.56	544	0.015	0	3.6	178	0.04	5.2
3	solid	SrTiO ₃	311	88	0.4	1.56	544	0.02	0.09	3.6	178	0.04	5.2
4	dashed	BaTiO ₃	2000	34	2.5	1.0	510	0.06	0	2.0	183	0.03	5.3
4	solid	BaTiO ₃	2000	34	1.7	1.0	510	0.025	0.21	2.0	183	0.03	5.3
5 ^b	solid	BaTiO ₃	2000	34	1.8	2.0	183	0.01	0.08	1.0	510	0.06	5.3
6	dashed	KTaO ₃	209	85.1	0.6	2.2	549	0.043	0	5	199	0.012	4.3
6	solid (4-34 μ)	KTaO ₃	209	85.1	0.5	2.3	549	0.040	0.12	5	199	0.012	4.3
6	solid (40-80 μ)	KTaO ₃	209	85.1	0.45	5.0	199	0.020	0.07	2.3	549	0.043	4.3
7		BaTiO ₃	same as Fig. 4—solid curve										

^a In cases where $\gamma_{12} = 0$, all modes are independent. The mode strength $4\pi z_1^2/\omega_1^2$ is the same as $4\pi\rho_1$ in the notation of Ref. 3.

^b The sign of $z_1 \cdot z_2$ must be taken as negative in all interaction calculations except for Fig. 5. For this case the interacting charges have the same sign.

their far infrared results.⁹ We have not attempted to fit their data but only note that the damping interaction is capable of giving the required dispersion effect near 180 cm^{-1} if $z_1 z_2$ is taken to be positive.

Miller and Spitzer have shown that the reflectivity of KTaO₃ can be fit exactly with the independent oscillator model by allowing the damping constant of the lowest frequency mode to change with frequency.¹⁰ The changes they require vary over two orders of magnitude. Our results cannot be directly compared with the Miller and Spitzer results because our Eq. (7) cannot be rewritten as the sum of independent oscillators with frequency-dependent damping constants. There is substantial agreement on the experiment problem. To fit the KTaO₃ reflectivity data, the over-all damping must be reduced in certain regions. Our thesis is that such reduction can be characterized by the simple model presented here and is due to an interference or competition during decay of the two (or more) optic phonons. It is not primarily due to details of the phonon spectrum.¹¹

In each case shown in the figures, it is the strong, lowest frequency mode which interacts with one of the other modes. The reason for this is that when the infrared signal has a frequency near one of the higher modes, the lowest frequency mode still vibrates with a very large amplitude even though it is far from resonance. If we consider the frequency to pass slowly through the higher mode resonance we have the ions

participating in each mode undergoing large displacements first in phase then a little later, 180° out of phase. Thus, there will be a great difference in the losses below and above resonance, if there is some anharmonicity in bonds connecting these ions.

The most striking feature of Eq. (7) for ϵ is the behavior of ϵ_1'' , the lossy part of the dielectric constant associated with the lowest frequency mode. Near ω_2 , ϵ_1'' goes through a dispersion and has a negative going spike over a small-frequency region (see Fig. 7). Thus, this mode by itself is giving power to the field. The over-all ϵ'' is, of course, positive, but the spike does cause the reduction in losses which is a feature of the

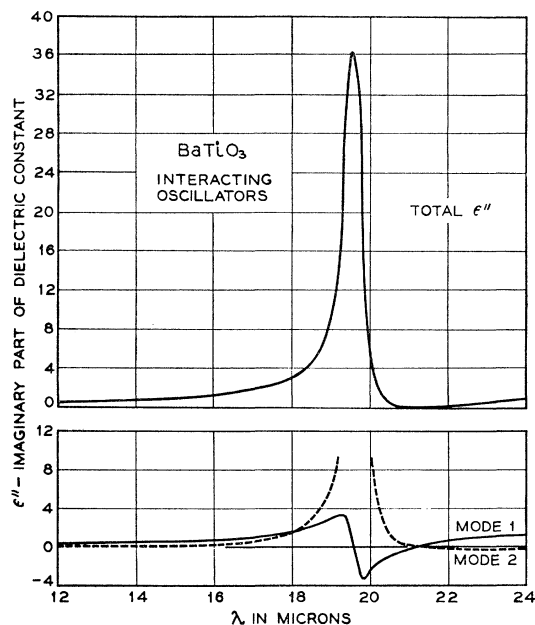


FIG. 7. Lossy part of the dielectric constant (ϵ'') for the interaction model fitted to BaTiO₃. The lower part of the figure shows ϵ'' for modes 1 and 2 separately. ϵ'' due to mode 3 (at 183 cm^{-1}) is too small to show.

⁹ S. Ikegami, I. Ueda, S. Kisaka, A. Mitsuishi, and H. Yoshinaga, J. Phys. Soc. Japan 17, 1210 (1962).

¹⁰ R. C. Miller and W. G. Spitzer, Phys. Rev. 129, 94 (1963). In this work the authors have neglected resonant-frequency shifts which are associated with the frequency-dependent damping. These shifts arise in a natural way in the theory (Ref. 5) and must be inserted in any phenomenological expression to satisfy the Kramers-Kronig relations.

¹¹ The transition probability expression given by Eq. (1) must be multiplied by a density of final states and summed over the final-state phonon spectrum. Deviations from the independent oscillator model can arise from the phonon spectrum therefore in addition to the deviations which arise from the coupling being considered here.

titanate spectrum. This effect is shown in Fig. 7 for BaTiO₃. In the lower part of the figure the contributions of the two interacting modes are shown separately. The dispersion in ϵ_1'' (which arises from the interaction damping) is seen to cause a reduction in the total ϵ'' on the low-frequency side of the mode. The effect is the same in SrTiO₃ and KTaO₃. The appearance of a negative ϵ_1'' does not violate any theorem on passive devices. The modes cannot be coupled to individually, so only the total field, total polarization, and total dielectric constant concern us. The total ϵ'' does remain positive.

Different models like those corresponding to Eqs. (4) and (5) will of course provide the same fit to data since they are equivalent within a unitary transformation. These models can be of use, however, in providing insights into the physical system of lattice vibrations. In this regard, it is particularly striking to note the results of transforming the parameters appropriate to the fits in Figs. 3, 4, and 6 (solid curves) using Eq. (5), to the parameters for Eq. (4) where the coupling is represented by a spring. Table II shows the force constant and damping matrices and the effective charge vectors for both models. We find that the equivalent two-mode system described by Eq. (4) (lower half of Table II) has one mode with all the damping (Γ_1) and nearly all the charge (e_1),¹² but coupled by a spring to an undamped mode. Viewed from this representation not only is it possible to fit the experimental curves better than with the independent oscillator model, but two of the otherwise arbitrary parameters are in fact approximately zero. Said in another fashion, we find using this viewpoint that it is the total polarization of the two modes which is heavily damped.

SUMMARY

Fits to the reflectivity of BaTiO₃, SrTiO₃, and KTaO₃ have been considerably improved by considering the interaction of optic modes in a crystal during the

¹² The squares of the effective charges should be compared since these determine the dielectric constant.

TABLE II. Force constants, damping factors, and effective charges for the best reflectivity fits. See Eqs. (4) and (5) for notation.

Diagonal force model				
		BaTiO ₃	SrTiO ₃	KTaO ₃
Force	ω_1^2	1156	7744	7242
	ω_2^2	260 000	296 000	301 000
Damping	$\gamma_1 + \gamma_{12}$	57.8	35.2	42.5
	$\gamma_2 + \gamma_{12}$	12.7	10.9	22.0
	γ_{12}	27.6	19.7	25.9
Charge	$(4\pi)^{1/2}z_1$	1520	1552	1230
	$(4\pi)^{1/2}z_2$	-510	-679	-833
Diagonal damping model				
		BaTiO ₃	SrTiO ₃	KTaO ₃
Force	$k_1 + k_{12}$	48 860	76 130	100 000
	$k_2 + k_{12}$	212 400	227 000	208 600
	k_{12}	100 400	122 600	136 700
Damping	Γ_1	70.9	46.2	60.2
	Γ_2	-0.4 ^a	-0.1 ^a	4.3
Charge	$(4\pi)^{1/2}e_1$	1592	1686	1485
	$(4\pi)^{1/2}e_2$	192	162	2.2

^a The small negative values for Γ_2 are the result of the original reflectivity fit. Very small changes of the original parameters (Table I) can cause Γ_2 to be zero or slightly positive without causing noticeable changes in the reflectivity. Theoretically, Γ_2 must, of course, be positive.

energy decay process. A simple classical model of the interaction provides in a quite natural way the reduction in damping at frequencies near certain optic modes. One parameter specifies the strength of the interaction. This parameter is the only additional quantity which must be determined in the usual process of fitting an infrared reflectivity spectrum. The interaction parameter may be represented in a mechanical model by a spring or a dashpot. Transformation to the spring-coupling representation has shown that for certain modes in the perovskites it is the total polarization which is heavily damped. The interaction process is very important in materials with one very strong mode and several weaker modes such as those presented here. It may be important in careful fits to any multimode crystal if variations of damping constants with frequency are to be studied.