Substituting this into Eq. (4) and integrating gives

$$n_T(t) = [kn_A(0)/(\beta_A + k - \beta_T)](e^{-\beta_T t} - e^{-(\beta_A + k)t}) \quad . \qquad (6)$$

Note that in both Refs. 1 and 2 direct activator excitation is considered negligible as has been assumed in deriving Eq. (6). Neglecting the coefficients of the exponential factors which are lost in normalizing the expressions, Eqs. (6) and (7) of Ref. 1 are exactly the same as Eqs. (5) and (6). The notation is simply changed as follows: $\beta_A \to a/\tau$, $\beta_T \to b/\tau$, $k \to (1-F)C/\tau$.

In summary, we feel that the random-walk calculations of Rosenstock are mathematically interesting, but they do not resolve the anomalies observed in the time evolution of the fluorescence intensities in tetracene-doped anthracene. Instead they give the same predictions as exciton diffusion theory, as should be expected. We conclude by emphasizing that due to the anomalous R_0 we do not feel that the results of Ref. 2 represent proof of the existence of long-range energy transfer in this system. They simply show that the current theories of energy transfer are not adequate for explaining phenomena occurring in short times and indicate the necessity for a time dependence similar to that of the long-range-interaction mechanism.

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PHYSICAL REVIEW B VOLUME 2, NUMBER 4 15 AUGUST 1970

uv Resonant Raman Scattering in ZnO

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Multiphonon scattering has been observed in ZnO by means of uv laser Raman studies. Phonon-scattering shifts greater than 4000 cm^{-1} are observed. Comparisons are made with other resonant Raman scattering studies, which now include the related series ZnTe, ZnSe, ZnO, and ZnS. A discussion of multiphonon linewidths is given.

Recently, resonant Raman scattering studies have been performed on a wide class of semiconductors, including CdS,[1] ZnSe,[2] InAs,[3] InSb,[4] ZnTe,[5] GaP,[6] and ZnS.[7] In the most recent paper,[7] it was observed that the number of multiphonon lines observed in III–V's and II–VI's at resonance varied monotonically with the polaron coupling coefficients in the materials examined. Since ZnO is known to have a large polaron coupling coefficient,[8] we have examined it in the present experiment as a comparison with earlier studies. The band gap in ZnO is at about 3400 Å, and so a uv laser is required as the excitation source for resonant scattering. We have employed a 1-m helium-cadmium laser of 4-mm bore and 3-Torr helium pressure, operating with 1 g of Cd isotope 114 in a sidearm at ~280 °C. The source emits 6 mW of cw power at the 3250-Å Cd II transition wavelength.

The spectrum obtained at ambient temperatures is shown in Fig. 1. Note the presence of emission lines from Cd I and II and from He I. Detection was by means of a Spex 1400 double monochromator, an EMI 6256 phototube with quartz window and collection optics, and a Keithley 610B electrometer.

The spectrum as shown consists of a broad luminescence envelope peaking at about 3750 Å, with relatively sharp lines superimposed upon it. These sharp lines are at frequency shifts which are multiples of the 1-LO zone-center frequency of 585 cm^{-1} and have been discussed in some detail for

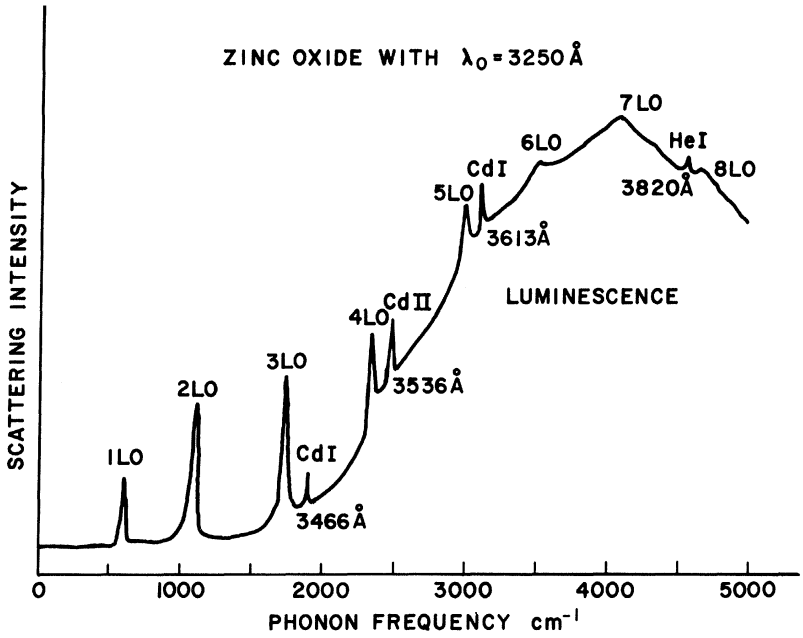

FIG. 1. Raman Spectrum of ZnO for $X(ZX)Y$ scattering at 295 °K and 3250-Å excitation.

other II–VI's elsewhere.[5,9]

Recently, we pointed out that the number of multiple phonon-scattering processes observed in semiconductors varies monotonically with polaron coupling coefficient,[7] from $n=2$ in InSb ($\alpha_e=0.02$) to $n=9$ in CdS ($\alpha_e=0.71$). In ZnO, which has a larger polaron coupling coefficient than CdS, we see only 8 LO, compared to 9 LO in CdS. However, we believe this reflects the slightly better sensitivity of the CdS experiment, which employed a higher laser power. We note that in an essentially identical experiment on CdS, Klein and Porto saw only 7 LO. Note that while only 8 LO are observed in ZnO, the larger phonon frequency (585 cm^{-1}) in that material yields enormous frequency shifts. Just as n(LO) can be compared with α, the polaron coefficient, the maximum frequency shift $n(\mathrm{LO})\times\omega_{\mathrm{LO}}$ can be compared with the deformation energy, which is equal to $\frac{1}{2}\alpha\hbar\omega_{\mathrm{LO}}$. Hence the large 8-LO shift reflects ZnO's large deformation energy.

Raman studies were also performed on ZnO using 4416-Å excitation from the same laser. Spectra obtained were essentially identical to those reported by Porto *et al.*[10] for 5145-Å excitation, i.e., they were nonresonant and included broad second-order features.

Finally, we would like to comment on the linewidths of the 6-LO, 7-LO, and 8-LO features in Fig. 1. The data shown were recorded with a spectral slit width of approximately 1.5 Å. At 3300 Å this is about 14 cm^{-1}. We believe that the widths greater than 14 cm^{-1} exhibited by the higher-order scattering processes are due to anisotropy and do not reflect lifetimes. This is explained below.

The 1-LO states have frequencies which vary[10] by 9 cm^{-1}, because of the anistropic short-range forces in the uniaxial ZnO lattice, varying from 583 cm^{-1} for polarization field $\mathcal{E}\perp c$, where c is the optic axis (an E_1 LO mode), to 574 cm^{-1} for $\mathcal{E}\parallel c$ (an A_1 LO mode).[10] The multiphonon scattering accentuates the effect of anisotropy. For example, the 2-LO frequencies should range from 2×574 cm$^{-1}=1148$ cm^{-1} to $2\times583=1166$ cm^{-1}, or, in general, the width of any multiphonon n-LO feature in ZnO should be equal to $9n$ cm^{-1}, as long as $9n$ cm^{-1} is larger than the one-phonon lifetime contribution to linewidth (about 3 cm^{-1} in ZnO at 300 °K) and also larger than the spectral slit width. All the ZnO multiphonon linewidths observed in this work fit the formula $\lambda(n\ \mathrm{LO})=9n$ cm^{-1} within the experimental uncertainty. Indeed, this is a probable reason for our failure to observe 9 LO and 10 LO.

In crystals such as CdS where the LO anisotropy is smaller (~3 cm^{-1}) the broadening of multiphonon features is smaller. However, even in cubic crystals it is anticipated that a formula of the form $\lambda(n\ \mathrm{LO})=na$ cm^{-1} will be valid if na cm^{-1} is larger than the one-phonon lifetime contribution and larger than the spectral slit width. For cubic crystals, a is not an anisotropy, but a dispersion parameter equal to the difference in frequency between LO at $K\approx10^6$ cm^{-1} and LO at $K\approx10^7$ cm^{-1}, the region of the Brillouin zone we believe participates

TABLE I. ZnO multiphonon frequencies (±3 cm^{-1}).

	(cm^{-1})
1 LO	585
2 LO	1165
3 LO	1749
4 LO	2343
5 LO	2928
6 LO	3520
7 LO	4101
8 LO	4678

in LO overtone scattering.[5] Since a is typically less than 1 cm^{-1} in cubic crystals, we ordinarily see[5] multiphonon linewidths dominated by one-phonon lifetimes or spectral slit width and, hence, approximately independent of n, the order of the scattering process.

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PHYSICAL REVIEW B VOLUME 2, NUMBER 4 15 AUGUST 1970

Sound Propagation near the Structural Phase Transition in Strontium Titanate†

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Finite ultrasonic velocity changes at the structural phase transition in $SrTiO_3$ are observed for different modes. They are interrelated and correlated by theory. No critical effects are observed.

Ultrasonic waves are an effective probe for studying static and dynamic aspects of phase transitions.[1] Here we would like to present ultrasonic velocity data near the structural phase transition in strontium titanate ($SrTiO_3$). We will interpret and correlate them with recent theories[2,3] and Brillouin scattering experiments.[4] Our results show that this phase transition exhibits characteristic features of a Landau-type displacive phase transition – namely, pronounced but finite velocity changes and the absence of critical effects, such as observed in magnetic phase transitions.[5] Our experiments supersede previous ones[6] in two important aspects: First, our transition occurs ~8 °K lower, probably due to improved crystal quality and, second, due to our special measuring technique,[7] we were able to measure through the transition region for various geometries, which enables us to interpret our data quantitatively.

It is now well established that the $T_a \sim 106$ °K phase transition in $SrTiO_3$ has associated with it a soft optic mode[8,9] and an order parameter ϕ which describes a static rotation of the oxygen octahedra.[10,11] If one also considers the strain–soft-mode interactions, then theory predicts a finite change in the elastic constants at T_a.[2,3,12] Taking the full symmetry of the crystal, theory[2,3,13] predicts for the elastic constants for $T < T_a$

$$c_{11} = c_{11}^c - 1.6D, \quad c_{33} = c_{11}^c - 4D, \quad c_{12} = c_{12}^c - 1.6D,$$

$$c_{13} = c_{12}^c + 2.4D, \quad c_{44} = c_{44}^c - E, \quad c_{66} = c_{44}^c \qquad (1)$$

(c_{ij}^c are cubic elastic constants); i.e., the elastic constant changes at T_a can be represented by two