anism consistent with existing data which can explain exponential edges in both ionic and covalent solids in a unified manner. The details of such a unified theory (and of the present calculation) will be presented in a subsequent paper[12] in which other types of theories of exponential edges will be considered and will be found to be incapable of the generality which this model provides. Experiments supporting and testing these arguments will be cited and suggested, respectively.

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$$T^* = (\hbar\Omega_0/2k_B)\coth(\hbar\Omega_0/2k_BT)$$

Here $\hbar\Omega_0$ is the energy of the longitudinal optical phonon.

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Imprisonment of Resonant Phonons Observed with a New Technique for the Detection of 10^{12}-Hz Phonons

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A new technique for the monochromatic detection of 10^{12}-Hz phonons is applied to the study of 20-cm^{-1} phonons in ruby. The resolution of the phonon detector in space and time allows the observation of the local imprisonment of 29-cm^{-1} phonons in a ruby crystal that contains variable concentrations of Cr^{3+} ions in the excited $\bar{E}(^2E)$ state. The application for the determination of the lifetime of 10^{12}-Hz phonons is discussed.

We report the direct observation of imprisonment of resonant phonons in ruby using a new technique for phonon detection. The frequency of the trapped phonons (29 cm^{-1} or 0.87×10^{12} Hz) corresponds to the separation of the $\bar{E}(^2E)$ and $2\bar{A}(^2\bar{E})$ levels of the Cr^{3+} ion in $\mathrm{Al_2O_3}$ [see Fig. 1(a)]. Transitions between these two excited states by the resonant phonons limit their mean free path severely and lead to the local imprisonment of these phonons reported here.

The resonant phonons are only scattered by *excited* Cr^{3+} ions, the number of which can easily be varied by changing the intensity of the exciting optical-pump radiation [see Fig. 1(a)]: Absorption of the pump light (from a mercury lamp) and subsequent radiationless transitions (RL) lead to the occupation of the metastable excited states $\bar{E}$ and $2\bar{A}$. The relative population of both levels is determined only by the 29-cm^{-1} pho-

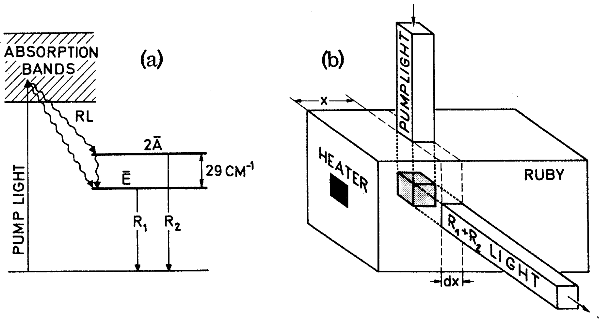

FIG. 1. Optical detector for 29-cm^{-1} phonons in ruby. (a) The energy levels of Cr^{3+} are shown schematically: The (blue and green) pump light is absorbed in the broad absorption bands. The wavelengths of the R_1 and R_2 fluorescence lines are 6935 and 6922 Å, respectively. (b) The experimental arrangement: The 50-Ω heater is evaporated on the 3×3-mm^2 surface of a 17-mm-long crystal (*a* direction). The detector volume (shaded has a size of about $1\times1\times1$ mm^3 and can be shifted relative to the heater.

nons and the corresponding equilibrium is established by one-phonon processes in a time shorter than 1 nsec.[1,2]

The population of the $2\bar{A}$ level (relative to the $\bar{E}$ level) is therefore a direct measure for the number of phonons at 29 cm^{-1}. For this reason we used the fluorescent intensity of the R_2 line (relative to that of the R_1 line) as a simple and instantaneous monitor of the monochromatic phonons within a small bandwidth ($\Delta\nu < 0.2$ cm^{-1}).

The experimental arrangement is shown schematically in Fig. 1(b). The ruby crystal (containing 0.05 mol% Cr^{3+} ions) was immersed in liquid helium at 4.2°K. The active detector volume itself corresponds to the common volume of the pump and fluorescence beams. Its size (about 1 mm^3) is small compared to the total crystal volume, and its location can be shifted by moving the crystal relative to the two light beams.

By observing the fluorescent light emitted from the active volume one can either observe the temporal changes of the phonon density at a fixed place or measure at a definite time the spatial distribution of the 29-cm^{-1} phonons within the crystal. We have studied 29-cm^{-1} phonons contained in the spectrum of repetitive heat pulses.[3,4] Of all the frequency components in a heat pulse only the 29-cm^{-1} phonons are monitored when traversing the detector volume. The intensity of the induced R_2 radiation J_{R_2}, which is very weak for a normal heat pulse (pulse duration 100 nsec, peak power 20 W), was isolated from the strong R_1 background by three narrow-band interference filters and detected by a low-noise photomultiplier using photon counting[5] and sampling technique: Signal pulses from the photomultiplier were counted only after passing an electronic gate triggered to open for a duration of 40 nsec but with variable delay after the heat pulses.

Figure 2 shows the measured temporal decay of the 29-cm^{-1} phonons in the detector volume located directly behind the heater for two different concentrations N^* of excited Cr^{3+} ions. The number of 29-cm^{-1} phonons drawn in Fig. 2 is given by the ratio J_{R_2}/N^*. For small N^* the observed phonon pulse [Fig. 2(a)] corresponds to the ballistic flight of a 100-nsec pulse of transverse phonons (velocity $v_T \approx 6\times10^5$ cm/sec) through the detector volume. If we raise N^* from its small value in Fig. 2(a) by increasing the optical-pump intensity by about two orders of magnitude, the phonons cannot escape so easily because of diffusion by resonant scattering and the decay time becomes remarkably lengthened [see Fig. 2(b)]. Our further experimental results indicate that the propagation of 29-cm^{-1} phonons is already seriously limited to the detector volume in the presence of only 10^{15}/cm^3 excited Cr^{3+}.

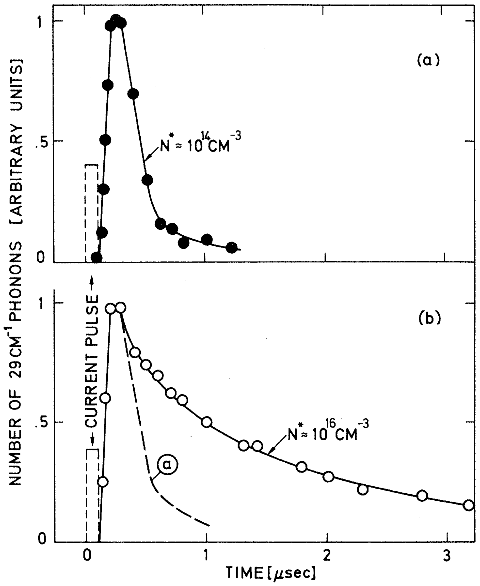

FIG. 2. 29-cm^{-1} phonons of a heat pulse observed in the detector volume located adjacent to the heater [$x=0$ in 1(b)]. The curve in (a) shows ballistic phonon propagation for the small number N^* of excited Cr^{3+} and the curve in (b) demonstrates phonon trapping when more Cr^{3+} are excited in the detector volume by stronger pump light. The time delay (100 nsec) between phonon and current pulse corresponds to signal delay in the photomultiplier.

We have also measured at a fixed delay time t_0 (measured after the heat pulse) the spatial phonon distribution in the crystal for the two cases of weak and strong optical pumping (see Fig. 3). In accordance with the results of Fig. 2 we find that for small N^* [Fig. 3(a)] the 29-cm^{-1} phonons propagate ballistically while for large N^* they cannot propagate appreciably [Fig. 3(b)]: They are imprisoned in a small volume directly behind the heater.

From further diffusion measurements we find that for the strongest optical pumping ($N^* \approx 10^{16}$ cm^{-3}) the mean free path for resonant scattering is of the order of 10^{-3} cm (in agreement with a theoretical estimate[6]). Because the corresponding time of flight (about 2×10^{-9} sec) of the 29-cm^{-1} phonons between their emission and reabsorption by excited Cr^{3+} is longer than the lifetime (3×10^{-10} sec) of the $2\bar{A}$ level for phonon emission,[2] we have the interesting case that the resonantly trapped 10^{12}-Hz phonons are contained

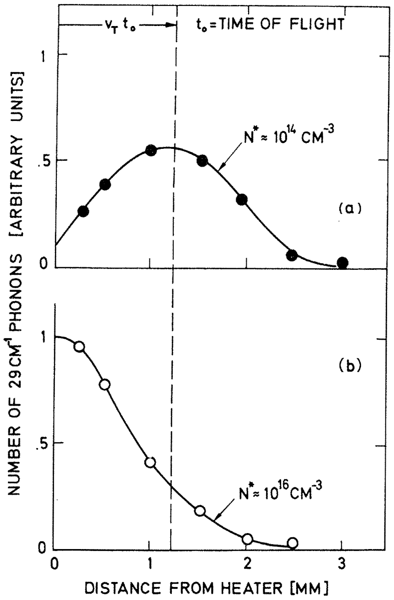

FIG. 3. Spatial distribution of 29-cm^{-1} phonons in the crystal 200 nsec after emission of the heat pulse. The width of the ballistic phonon pulse in (a) is larger than the 0.6 mm expected from the duration of the heat pulse (100 nsec) due to the dimension of the detector volume [dx = 1 mm in Fig. 1(b)]. The curve in (b) indicates the imprisonment of 29-cm^{-1} phonons directly behind the heater when the crystal contains a larger concentration of excited Cr^{3+}.

in the crystal as phonons, in contrast to the systems studied up to now for which trapped resonant radiation is stored mainly as electronic excitation energy (in case of trapped bottleneck phonons[6,7] as well as in case of trapped resonant photons[8,9]).

Therefore, we can determine the lifetime τ_{ph} of the 10^{12}-Hz phonons from the exponential decay of the imprisoned phonons [Fig. 2(b)]. The resulting time, $\tau_{\text{ph}} \approx 1.5$ μsec, corresponds to the spontaneous decay time of the 29-cm^{-1} phonons in ruby because no temperature dependence between 4.2 and 2°K has been observed.

Applications of the described detection principle for 10^{12}-Hz phonons are in preparation for other crystal-impurity systems. By applying external fields to the crystal it should be possible to realize tunable detectors for 10^{12}-Hz phonons, thus extending the region of existing phonon spectrometers.[10]

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Observation of Simultaneous Excitation of the Coupled Cr^{3+} Ions to the ^2E_g States in LaAlO$_3$:Cr^{3+}

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Transitions to the states of Cr^{3+} pairs where both ions are excited to the ^2E_g levels have been observed in LaAlO$_3$:Cr^{3+}. Of the predicted eight pair levels, three of the $S=0$ and one of the $S=1$ levels have definitely been identified at approximately twice the ^2E_g energy absorption. The remaining transitions either are obscured by the strong vibronic spectrum or are very weak. The polarization of the lines agrees with a model based on the mechanism of exchange-induced electric dipole moment.

Previous studies of exchange-coupled Cr^{3+} pairs have been concerned with the spectra resulting from transitions between the pair levels where both ions are in the $^4A_2$ ground state and levels where one of the ions is in the ^2E state and the other in the $^4A_2$ state.[1] We report the experimental observation of a sharp line spectrum of Cr^{3+} pairs in LaAlO$_3$ resulting from transitions to levels where both ions of the pair are in the ^2E_g state. The levels are found at approximately