Having ruled out the Γ, L, and X transitions on the basis of the parity argument, we explore the noncentrosymmetric points in the Brillouin zone. Based on the band-structure calculation of Onodera, Okazaki, and Inui,[10] we may rule out Σ, K, S, and Λ as unsuitable on the basis of the energy separation and the density-of-states argument. Furthermore, angular functions associated with these points which describe the polarization data would have noticable dips. For example, the Λ-point transitions are described by three angular functions,[13] and all three functions predict dips in our polarization measurement when $\alpha = 32°$ and $58°$.

The most likely candidate which could explain this structure is the Δ transition. Of the five angular functions,[13] the A_1 function fits our polarization data within the experimental uncertainty. This results in the choice of $\Delta_6 \rightarrow \Delta_6$, since it is the only transition that contains the A_1 function.

In conclusion, we tentatively assign the peak at 7.21 eV to the $\Delta_6 \rightarrow \Delta_6$ transition on the basis of the parity and polarization arguments. One of the important results of this assignment is that the spin-orbit splitting of the valence band must be smaller than the 1.26 eV calculated by Onodera, Okazaki, and Inui.[10] In fact, Phillips's assertion of 0.6 eV, based on the free-iodine splitting parameter, seems much more plausible. We intend to pursue this point further when our new experimental arrangement is completed, which will allow us to investigate the spectrum up to 9.2 eV. The peak at 7.375 eV does belong to a centrosymmetric point by the parity argument.[11] The polarization dependence of this peak cannot be measured at the present time due to the severe limitation of the uv light. This point also will be studied in the new experimental arrangement.

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EXCITON SPECTRA OF CaO AND MgO†

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Exciton spectra of crystalline CaO and MgO at 25°K are presented and interpreted in terms of valence-band spin-orbit splitting and strong exciton-phonon coupling.

Because of the current interest in the exciton spectra of II-VI compounds, we have measured the reflectance spectra of CaO and MgO at higher resolution and lower temperature than those reported by Glasscock and Hensley[1] and Roessler and Walker.[2] New fine structure in both spectra was observed in the present work.

Measurements were made on single crystals which were cleaved and mounted in a dry nitrogen atmosphere to reduce surface contamination. The measurements were made on a McPherson 225 monochromator equipped with an ultrahigh-vacuum, liquid-helium reflectometer.

The exciton spectrum of CaO was obtained with an instrumental resolution of better than 0.006 eV in the region of the molecular-hydrogen continuum at 300, 80, and 25°K. A Kramers-Kronig analysis of the low-temperature spectra augmented by room-temperature measurements from 6 to 30 eV was performed. Results from CaO at

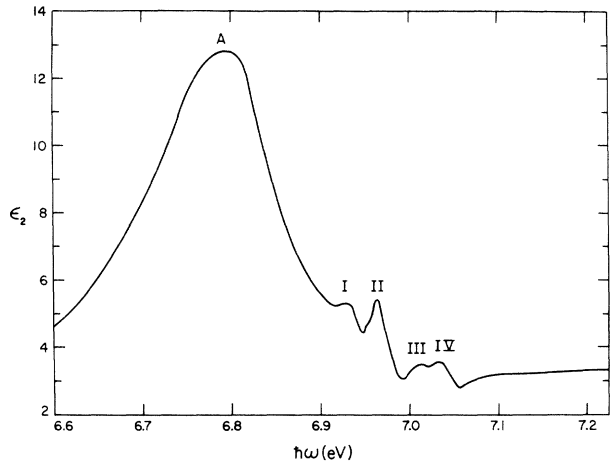

FIG. 1. Imaginary part, ϵ_2, of the dielectric constant of CaO at 25°K.

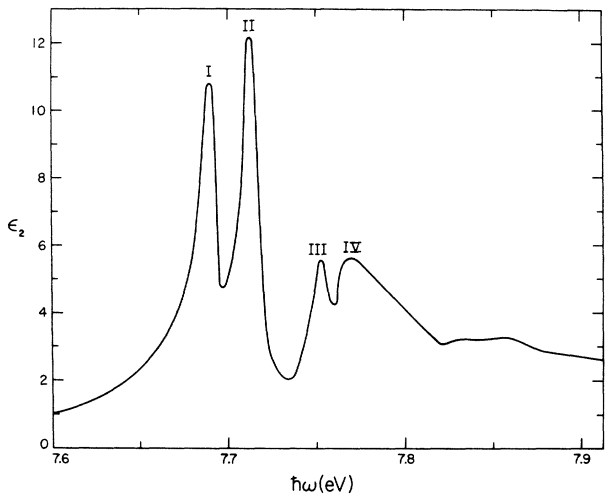

FIG. 2. Imaginary part, ϵ_2, of the dielectric constant of MgO at 25°K.

25°K are shown in Fig. 1.

The Γ exciton of MgO was studied at 80 and 25°K using an instrumental resolution of about 0.003 eV in the region of the line spectrum of molecular hydrogen from 7.6 to 7.9 eV which gave about three times the data-point density obtained previously.[3] An ϵ_2 spectrum of MgO at 25°K is shown in Fig. 2.

For both CaO and MgO two doublets occur. In CaO the doublets, which are not visible at 300°K, arise out of the background at 80°K. From 80 to 25°K the two doublets sharpen slightly and shift to higher energy. The positions of the four peaks in both reflectance and ϵ_2 at 25°K are shown in Table I along with the separations between peaks I and II and between III and IV (labeled $\delta E_{\text{S.O.}}$), and the separations between peaks I and III and between II and IV (labeled δE_{EPC}).

We believe that in both crystals the first doublet corresponds to a spin-orbit–split Γ exciton and the second doublet corresponds to spin-orbit–split exciton-phonon complex (EPC). The relative oscillator strength of the second doublet is too large to be accounted for solely by the $n=2$ state of the exciton. Liang and Yoffe[3] have suggested the existence of an EPC in MgO based on earlier MgO data.[2] The energy separations of the two peaks in the doublets, shown in Table I as $\delta E_{\text{S.O.}}$, are close to the value of 25 meV predicted[4,5] for the spin-orbit splitting of the valence band obtained from the O^- ion; however, the splitting of the EPC is somewhat less than the spin-orbit splitting of the Γ exciton. The energy separation of the Γ excitons from the position of the EPC's (δE_{EPC}) shown in Table I are close to the values of 89 and 70 meV of the $k=0$ LO-phonon energy in MgO[6] and CaO,[7,8] respectively.

Toyozawa and Hermanson[9] have pointed out that the discrepancy between the energy of an EPC and lowest order perturbation theory for simultaneous phonon emission is $(\Delta+\gamma)\omega_0$, where ω_0 is the LO-phonon energy, γ is a coupling constant of the order of unity, and $\Delta=(\omega_0-\delta E_{\text{EPC}})/\omega_0$. The experimental values of Δ for the $j=\frac{3}{2}$ Γ exciton and its EPC at 25°K are 0.30 and −0.20 for

Table I. Exciton fine structure of MgO and CaO. Exciton peaks, spin-orbit splittings ($\delta E_{\text{S.O.}}$), and exciton-phonon complex splittings (δE_{EPC}) are shown.

	Exciton peaks (eV)				$\delta E_{\text{S.O.}}$ (meV)		δE_{EPC} (meV)	
	I	II	III	IV	(I-II)	(III-IV)	(I-III)	(II-IV)
				MgO (25°K)				
R	7.689	7.715	7.752	7.768	26	16	63	53
ϵ_2	7.690	7.712	7.752	7.770	22	18	62	58
				CaO (25°K)				
R	6.939	6.973	7.012	7.034	34	22	73	61
ϵ_2	6.927	6.963	7.011	7.031	36	20	84	71

MgO and CaO, respectively. Although δE_{EPC} for CaO is slightly greater than ω_0, it is less than $(1+\gamma)\omega_0$ unless $\gamma \leq 0.2$. A band gap at Γ for MgO of 7.775 ± 0.010 eV is consistent with having four Lorentzian shaped peaks and a band edge rising as $E^{1/2}$. A Γ band-gap energy for CaO of 7.034 ± 0.022 eV was estimated as being between the high-energy end of the first and second EPC. From the Γ band gaps we computed the $j = \frac{3}{2}$ exciton binding energies ϵ_B of 85 ± 10 and 104 ± 22 meV for MgO and CaO, respectively. The value of ϵ_B/ω_0 is 0.95 ± 0.11 for MgO and 1.5 ± 0.3 for CaO.

A hydrogenic fit to the measured binding energy of MgO, based on the effective mass value of $\mu = 0.23m$ of Cohen and Fong,[10] can be obtained only by using an effective dielectric constant $\epsilon = 6.0$ intermediate between the optical $\epsilon_0 = 2.97$ and the static dielectric constant $\epsilon_\infty = 9.8$.

The intensity ratio of the oscillator strength of the EPC to that of the Γ exciton is 0.63 ± 0.07 for MgO and 0.40 ± 0.20 for CaO. Toyozawa and Hermanson[9] predict that the intensity ratio increases with ϵ_B/ω_0. Our two experimental data points for MgO and CaO do not support this feature of the theory.

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PHONON AVALANCHE IN Ce-DOPED LANTHANUM MAGNESIUM DOUBLE NITRATE. MEASUREMENT OF THE PHONON LIFETIME AND OF THE ACOUSTIC SPECTRUM

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We have investigated the phonon avalanche in Ce-doped lanthanum magnesium double nitrate using pulsed microwave and pulsed magnetic-field techniques. In these experiments we have been able to determine the lifetime of the avalanche phonons and to obtain some evidence for coherence in the spin-phonon interaction.

We have investigated the phonon avalanche in Ce-doped lanthanum magnesium double nitrate[1,2] (LMN-Ce) by means of electron spin-echo and pulsed magnetic-field techniques. The experiments were conducted at 9.1 Gc/sec and at 1.5°K on a nominally 0.3% doped sample of dimensions 0.2 cm × 0.3 cm × 0.3 cm. The resonance line had a full width at half-height of 10 G. Microwave pulses were generated by a Litton L5022 traveling wave tube (output $\gtrsim 1$ kW). Pulsed magnetic fields were obtained by winding a few turns of wire around the sample. In these experiments we have been able to determine the phonon lifetime in the sample. We have also been able to obtain some information regarding the form of the acoustic pulses which develop during the avalanche.

The phonon lifetime was investigated by using a three-pulse sequence as follows. Pulse I, a 180° pulse, inverts the Ce-spin system and operates effectively on the whole resonance line. Pulses II and III generate a two-pulse spin echo by means of which we sample the magnetization a time t after the inverting pulse. Since we wish to sample only the central portion of the line (i.e., the portion which is "burnt out" by the avalanche), pulses II and III are characterized by a lower microwave field H_1 than pulse I.[3] The echo signal is integrated by means of a boxcar unit, and its amplitude is traced out as a function of t as shown in Fig. 1.

The lowest curve in Fig. 1 (curve a) shows the normal evolution of the inverted magnetization as the avalanche proceeds, and is similar to the curve shown in Fig. 1 of Ref. 1. In obtaining the upper curves we interrupted the growth of the avalanche by switching the Zeeman field 15 G to one side of its initial value at a time $\simeq 8$ μsec after the inverting pulse. As a result of this sudden change in field the hot phonons were left in