higher than the 1.62 kHz per spin predicted by the restricted-trace approximation.

We are much indebted to Professor A. Abragam for his constant interest in this work.

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Radiative Decay of Core Excitons in Alkali Halides*

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The soft x-ray emission spectra for fresh films of LiF, LiCl, and LiBr are presented. Prominent structures in these spectra above the Li^+ 1*s* emission edge are shown to give a direct observation of the radiative decay of *K*-shell cation excitons in these materials.

The possibility of the existence of excitons associated with core electron states has stimulated several studies on ionic compounds; in particular on alkali halides. Extensive observations have been made of soft x-ray absorption spectra, with conflicting interpretations. These data and interpretations have been summarized and discussed by Gudat, Kunz, and Petersen,[1] by Pantelides and Brown,[2] and by Pantelides.[3] Structure in x-ray absorption spectra corresponds to the excitation of electrons from a flat core level to an exciton state or to the conduction band. In the absence of excitons, structure in x-ray absorption spectra should thus be proportional to the conduction-band density of states. The existence of a strong electron-hole interaction not only introduces corresponding structure into the absorption spectrum but modifies the core-level to conduction-band spectrum. On the basis of band-structure calculations modified by electron-hole interactions it has not been possible to identify any structure in the soft x-ray absorption spectrum of an ionic compound as being associated unambiguously with core-level excitons. Recently,[1-3] however, it has become generally accepted that some structures in the x-ray absorption spectra of some ionic compounds are in fact due to core-level excitons. This has resulted from the use of additional independent experimental data for the same material and not from the results of theoretical calculations based solely on the crystal structure. X-ray photoelectron spectra and optical-gap data were used to determine the core-level to conduction-band absorption threshold for each material. Structure appearing in the spectrum below this threshold was then identified as due to core-level excitons, i. e., bound states of the electron to the core hole.

We can show that corresponding structure in the x-ray emission spectra of the lithium halides can be identified positively as due to the radiative decay of excitons. Structure in x-ray emission spectra corresponds to electronic transitions either from the valence band or from excited states into holes in the core level. The probability of an electron excited to the conduction band undergoing radiative decay in this way is extremely small. However, an excited electron bound by the electron-hole interaction is held physically close to the hole and in some cases the probability of radiative decay is appreciable. Electronic transitions from the valence band to a core level may involve double ionization, resulting in structure above the valence-band emission edge. In the case of the lithium halides reported on in this Letter such transitions can be distinguished from transitions involving excitons by the magnitude of the energies involved. Thus some structure seen in x-ray emission spectra within the optical-gap energy above the valence-band emission edge can be attributed unambiguously to the radiative decay of core-level excitons. Maiste, Saar, and Élango[4] have reported the soft x-ray emission spectrum obtained from a sample of LiF powder ground into the grooved surface of a copper plate, and identified some structure in this spectrum as being associated with the radiative decay of a core-level exciton. More recently[5] the same experimental group has reported similar data for other ionic compounds. However, it is well known[6, 7] that the x-ray emission spectra of halogen-containing compounds change rapidly with time from the beginning of electron bombardment, the rate of change depending on the energy and intensity of the bombarding electron beam. Thus the x-ray emission spectra of alkali halides presented so far in the literature[4,5,7] do not correspond to the pure ionic compounds, but to more complicated systems.

In Fig. 1 we present, for the first time, the soft x-ray emission spectra for the pure alkali halides LiF, LiCl, and LiBr. The experimental apparatus used to obtain these spectra has been described previously.[8,9] Electrons from an electron gun (2 kV; $\frac{1}{2}$–1 mA/cm^2) were incident on films of LiF, LiCl, and LiBr. The photons emitted were dispersed by a soft x-ray scanning monochromator and then detected in the first order using a channel electron multiplier from which the output current was read with a logarithmic electrometer. The alkali halide samples were vacuum evaporated *in situ* directly onto the water-cooled copper anode for a sufficient time that the films were effectively infinitely thick. In certain regions of the spectrum where the intensity was found to change, even with a few seconds of electron bombardment, the monochromator was not used in its scanning mode but, with the monochromator set to record a given photon energy, a film of LiF was evaporated *in situ* and the photon intensity recorded as a function of time after the electron bombardment was started. The intensity at time zero was obtained by extrapolating this time-dependent curve. This process was repeated at different photon energies until the whole spectrum for LiF, presented in Fig. 1, had been obtained. This spectrum corresponds to that for pure LiF as each point on the spectrum corresponds to a fresh LiF film. The results for LiCl and LiBr were obtained in the same way. The pressure in the chamber was (3–5)× 10^{-7} Torr and the resolution in the x-ray spectra was limited only by the slit width of 600 μm for LiF, 400 μm for LiCl, and 300 μm for LiBr. This gave spectral slit widths in eV ranging from 0.008 at 42 eV to 0.75 at 140 eV for LiCl.

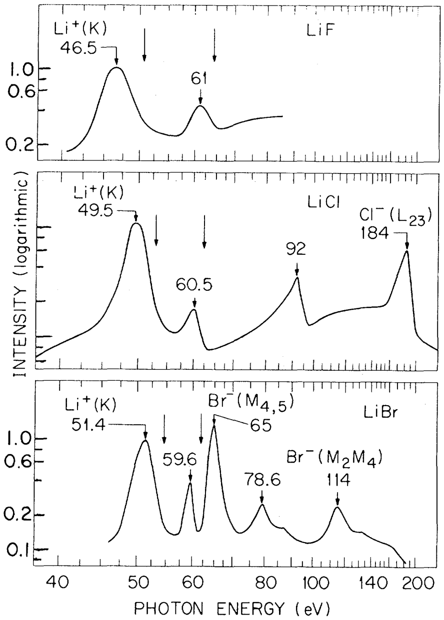

FIG. 1. Soft x-ray emission spectra from fresh surfaces of evaporated films of LiF, LiCl, and LiBr. For each alkali halide arrows mark the top of the valence band and the bottom of the conduction band as estimated by Pantelides and Brown (Ref. 2) from x-ray photoelectron spectra and optical-gap data.

From various observations, including independently measured x-ray photoelectron spectra and optical-gap data, it has been determined, as outlined by Pantelides,[3] that in LiF the top of the valence band lies 50.8 eV, and the bottom of the conduction band 64.4 eV, above the Li$^+$ 1s level. It is seen that the value for the top of the valence band is consistent with our emission spectrum for LiF shown in Fig. 1. Emission seen in the region of the bandgap must be excitonic in origin, since the probability of a free electron in the conduction band undergoing radiative decay to the Li$^+$ 1s level is negligible. Thus the emission seen at 61.0 eV must represent a direct observation of the radiative decay of a core-level exciton in pure LiF. Maiste and co-workers[5] have calculated the radiative lifetime for the lowest-energy exciton associated with the Li$^+$ 1s level in lithium halides as being between 10^{-11} and 10^{-12} sec. This reflects the fact that the K-shell cation excitons in lithium halides are highly localized,[10] so that the probability of radiative decay of cation excitons is high in these materials. In contrast, an electron excited to the conduction band would have to decay radiatively back into the hole within a time of $\lesssim 10^{-15}$ sec, i.e., before it has a chance of moving away from the vicinity of the Li^{++} cation. In fact, if it does stay in the vicinity of the Li^{++} cation it is more probable that it will decay initially to the exciton level than directly to the core level. We have considered but ruled out double ionization as the origin of the emission seen at 61.0 eV. We have shown experimentally[9] that double ionization in metallic lithium produces a high-energy satellite 28.5 eV above the K emission peak. Since the valence electrons reside almost exclusively on the halogen ions, it is anticipated that double ionization of the Li ion in LiF would not differ significantly from that in metallic Li and would result in an emission peak at ~75 eV, i.e., ~28.5 eV above the observed Li^+ K emission peak at 46.5 eV. Double ionization could also involve simultaneous ionization of nearest-neighbor Li $1s$ and F $2s$ levels. However, because of the physical separation of the core states in the Li and F ions the perturbation in this case on the Li K emission is much smaller than the observed 10.2 eV in our spectrum. The change in the potential energy of the Li $1s$ level due to the hole in the F $2s$ level would be ~7 eV in the absence of screening. With screening taken into account, the shift in energy is $\ll 7$ eV. On the other hand, once the peak at 61.0 eV is shown to be excitonic in orgin, the energies involved can be derived theoretically.[1-3,5]

Calculations[2,3] identical to those for LiF have set the top of the valence band at 51.9 and 54.2 eV and the bottom of the conduction band at 62.3 and 61.8 eV in LiCl and LiBr, respectively. The emission peaks seen in Fig. 1 at 60.5 eV for LiCl and 59.6 eV for LiBr are identified, by the same arguments as were used for LiF, as due to the radiative decay of core-level excitons.

The progression of exciton peak energies seen in our emission spectra (i.e., 61.0 eV in LiF, 60.5 eV in LiCl, and 59.6 eV in LiBr) is very similar to the variation of peak energies seen in the absorption spectra discussed by Pantelides.[3] This is shown in Fig. 2 where exciton peak energies observed in absorption and emission are compared with the conduction-band absorption thresholds. The peaks in emission are seen to be systematically lower in energy than in absorption as expected, because of rearrangement of the electrons around the core hole. The similarity of the positions of the exciton peaks in emission and absorption spectra, shown in Fig. 2, now gives direct experimental proof of the assignment of exciton absorption given to the peaks in the absorption spectra,[1-3] the same exciton states being involved in both absorption and emission. It might be pointed out that although we have quoted the location of the top of the valence band for each spectrum from Ref. 2, this is not an integral part of our argument. Our spectra, shown in Fig. 1, indicate approximate values for the tops of the valence bands and in each case the exciton emission is at a higher energy by several eV, is clearly resolved from the valence-band emission, and is definitely less than the optical-gap energy.

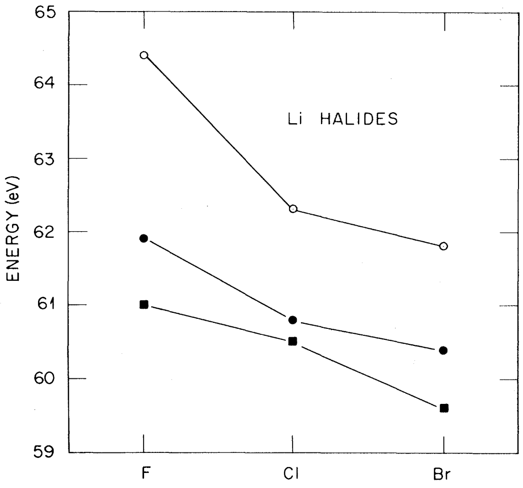

FIG. 2. Positions of the experimentally observed core-level exciton peaks in the Li^+ K emission spectra (■) taken from Fig. 1, compared with the experimentally observed core-level exciton peaks in the Li^+ K absorption spectra (●) and the position of the conduction-band thresholds (○) both reported in Ref. 3.

The emission spectrum for LiF shown in Fig. 1 should be compared with that obtained previously[7] using the same experimental equipment in the scanning mode. In the investigations reported in Ref. 7, crystalline LiF was cleaved, mounted in the apparatus, and bombarded with electrons. The soft x-ray emission spectrum was scanned continuously with photon energy. Since it took 1 to 2 min to obtain a whole spectrum, and the specimen was observed to change on electron bombardment such that within a few minutes the surface looked metallic, it is obvious that even the earliest recorded spectrum after the start of electron bombardment could not correspond to pure LiF. The excitonic radiative decay at 61.0

eV is barely evident in this case. When we bombarded an evaporated film of LiF and scanned the spectrum, as was done for the cleaved LiF in Ref. 7, the specimen surface appeared metallic within a few minutes after the start of electron bombardment, the main $K\alpha$ emission peak shifted from that of Li^+ in LiF at 46.5 eV to that for pure Li at 54.3 eV,[9] the exciton peak at 61.0 eV disappeared, and later emission due to contamination buildup appeared at ~50 eV.

Other emission peaks seen in the LiCl spectrum in Fig. 1 correspond to Li^+ in LiCl at 49.5 eV ($K\alpha$ emission) and Cl^- in LiCl at 184 eV ($L_{23}M_1$) with a second-order peak at 92 eV. The peaks in the LiBr spectrum correspond to Li^+ in LiBr at 51.4 eV ($K\alpha$ emission) and Br^- in LiBr at 65 eV ($M_{4,5}$) and 114 eV (M_2M_4). The peak at 78.6 eV in LiBr may be the Br^- M_1M_{23} doublet; however, a difference of a few eV from the tabulated values[11] is noted.

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COMMENTS

Massive-Particle Production in Neutrino Scattering*

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It is found that the transverse momentum spectrum of the nonleading muon in dimuon events is consistent with their originating in the decay of a massive particle.

The recent discovery of dimuons in high-energy neutrino scattering[1] has led to a great deal of speculation as to their origin.[2,3] I show that these events can be caused by the three-body decay of a particle of mass ~2.5 GeV.

According to the charm hypothesis, charmed pseudoscalar mesons of mass ~1.8 GeV should exist and be created, at least some of the time, in deep inelastic neutrino scattering. These are expected to decay promptly. Typical decay modes are

$$F^+ \to \mu^+\nu, \tag{1a}$$

$$F^+ \to \bar{K}^0\mu^+\nu. \tag{1b}$$

For Process (1a), in the rest frame of the F^+, the muon has a monochromatic energy spectrum

$$|\vec{P}_\mu| = (M_F{}^2 - m_\mu{}^2)/2M_F. \tag{2a}$$

$$E_\mu = (M_F{}^2 + m_\mu{}^2)/2M_F. \tag{2b}$$