by the acoustic band of the host lattice. This absorption appears to bear some relation to the "structures due to host-lattice dynamics," which are predicted by Benedek and Nardelli,[11] but is quite different from the low-frequency resonance mode which is the subject of most of Ref. 11. We only feel justified in describing these features as a selection of the delocalized acoustic-type modes of the impurity-distorted lattice. The *decrease* of the frequency of the lowest energy band with increasing pressure as potassium and rubidium halides approach their phase change is an indication of a tendency towards instability of the relevant modes.

In-Gap Localized Modes

For NaCl, KCl, RbCl, RbBr, RbI, CsCl, CsBr, and CsI, no gap modes have been observed for any ion at any pressure. These observations are in general agreement with calculations[24] which indicate that some of these halides do not possess an energy gap, and in those cases where no other experimental or theoretical evidence is available this work can be interpreted as showing that in these cases no energy gap exists.

Gap modes are observed for NaBr, NaI, KBr, and KI. Neutron-scattering studies have shown the existence of the energy gaps in NaI, KBr, and KI, and the observed gap modes are found to be located fairly centrally within these gaps. Karo and Hardy[24] have calculated that an energy gap exists in NaBr, but our gap modes (117–127 cm^{-1} for NCO^- and 110–123 cm^{-1} for CN^-) suggest that the gap is wider and at a higher energy than that calculated by them.

Torsional motion of the impurity ion seems to play an important part in most of the gap modes found for polyatomic impurity ions.

The gap modes in KBr and KI are not replaced by any equivalent structure when these halides are compressed into their high-pressure (CsCl-type) structure. No gap modes are found in the high-pressure external-mode spectra of the rubidium halides. Thus we feel confident in stating that there are no energy gaps in these materials in their high-pressure phases.

The pressure dependence of the gap-mode frequencies, especially for KBr, where the gap is very narrow, has been used as a guide to the pressure dependence of the bordering host-lattice frequencies.

External Modes within the Optic Bands of the Host Lattice

With only one exception, discussed below, only relatively broad features are found in the external-mode sidestructure in the energies covered by the host-lattice optic bands. These broad external-mode features have been used to indicate the extent of the host-lattice band, but their detailed interpretation clearly requires a refined theoretical investigation.

The one band under this heading which was found to become very sharp at very low temperatures is the 154-cm^{-1} (at 100°K) band in ν_{ext} for CN^- in NaBr. No explanation for this band is suggested, but it is seen as highly significant that this band is found, for the ⟨001⟩-oriented CN^-, at a frequency which corresponds to a marked low-absorption region for the ⟨111⟩-oriented NCO^-.

Superoptic Local Modes

For the various polyatomic-impurity ions which we have studied, these modes, if they are found to exist, always seem to be interpretable in terms of a torsional motion of the impurity. In many cases, one of these modes is found to be very close, and possibly coincident with, the highest optic-mode frequency of the pure lattice. Superoptic modes, if they are to exist at all, must be quite highly localized. One requirement for their existence is therefore that the nearest neighbors alone must be sufficiently massive to represent an adequate counterbalance (stationary center of gravity and zero net angular momentum) without themselves having too large an amplitude, thus involving another shell of ions. The failure of the six nearest-neighbor sodium ions to comply with the above condition is considered to be the main reason why no such modes have been found for our relatively massive impurities in the sodium halides.

Mainly the lattice-sideband structures shown in Figs. 7 and 8 have been used in the above discussion, but complementary information for other ions (see, e.g., Refs. 3–6) could be included to substantiate the above conclusions and in some cases give closer limits on phonon band gap energies and high-energy maxima of optic bands.

Although most of the lattice-sideband spectra which have been discussed have been due to impurity ions in alkali halides, some of the wider applications of this type of spectrum, and its pressure dependence, have been indicated.

ACKNOWLEDGMENTS

We are grateful to Professor W. C. Price, F. R. S., for laboratory facilities and encouragement. One of us (M. A. C.) is indebted to the Science Research Council for a maintenance grant, and we are grateful for financial assistance from the U. S. Army (Contract No. DA-91-591-EUC) and the Institute of Petroleum Hydrocarbon Research Group.