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Determination of the origin of the 10.6- μm absorption in CO_2 laser window materials

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Theoretical studies are presented which show that it should be possible to determine whether the absorption at 10.6 μm in alkali halides is due to impurities or whether it is intrinsic. These studies show that the temperature dependence of the measured absorption should be very different for intrinsic multiphonon absorption as compared with defect-activated absorption.

The development of high-power infrared lasers has led to a need for window materials having a very low absorption coefficient in order to avoid thermal lensing.¹ In the important 10.6 μm region, the alkali halides KCl and KBr appear to have the lowest absorption coefficients.² It has been suggested that the residual absorption in alkali halides at 10.6 μm may be impurity limited rather than intrinsic in origin. However, there is no easy experimental method of distinguishing between intrinsic and impurity absorption at these low levels. The purpose of this letter is to point out that some clue as to the origin of this absorption might be obtained by studying the temperature dependence of the absorption.

If the 10.6 μm absorption in KCl is due to higher-order lattice absorption, then it must be at least a five-phonon process. Such processes can be brought about either by the sixth-order anharmonic terms in the lattice potential energy or by the fifth-order dipole moment.³ In either case, the temperature dependence is the same. Thus we shall make the hypothesis that the intrinsic absorption involves the creation of five phonons, each of which has the same characteristic energy, and that this energy is given by $\frac{1}{5}\hbar\Omega$, where Ω is the angular frequency of the 10.6 μm radiation in free space. This is obviously an approximation, and more detailed investigation may well modify the contention that the five phonons have the same energy. However, examination of the single-phonon density of states for KCl⁴ indicates that the frequency calculated in this way lies in the region where the density of states is significant. Thus, since the selection rules for five-phonon processes are much less stringent than those for one- and two-phonon processes, it is reasonable to assume that the most probable five-phonon processes are indeed those for which the phonons in question have approximately the same frequency. Given this assumption, a computation of the temperature-weighting factor for either the anharmonic process or the fifth-order dipole moment process gives the following result:

$$A \propto (n_0 + 1)^5 - n_0^5, \quad (1)$$

where A is the absorption and $n_0 = [\exp(\hbar\Omega/5kT) - 1]^{-1}$, k is Boltzmann's constant, and T is the temperature. From this expression we have computed the expected temperature dependence, and the resultant curve is shown in Fig. 1 for values of T between 0 and 800 $^\circ\text{K}$.

If we now turn to the case where the absorption is defect activated, then from quite general considerations one

can argue that the absorption cross section for any given defect-induced absorption must be directly proportional to the number of defects. Since this is the case, it follows immediately that, unless the defect has an effective charge that is orders of magnitude greater than that of the ions of the host lattice, any 10.6 μm absorption due to the fifth harmonic of defect-activated lattice absorption will be completely negligible compared with the intrinsic 10.6 μm absorption. This is so because the impurity concentrations in KCl specimens that have been studied are reported as being in the parts-per-million range. Consequently, the defect-activated absorption will be $\sim 10^{-6}$ times smaller than the intrinsic absorption.

Thus we are forced to the conclusion that if the residual absorption at 10.6 μm is associated with an impurity mode, then it must be an impurity mode whose frequency lies sufficiently close to 10.6 μm for either the one- or

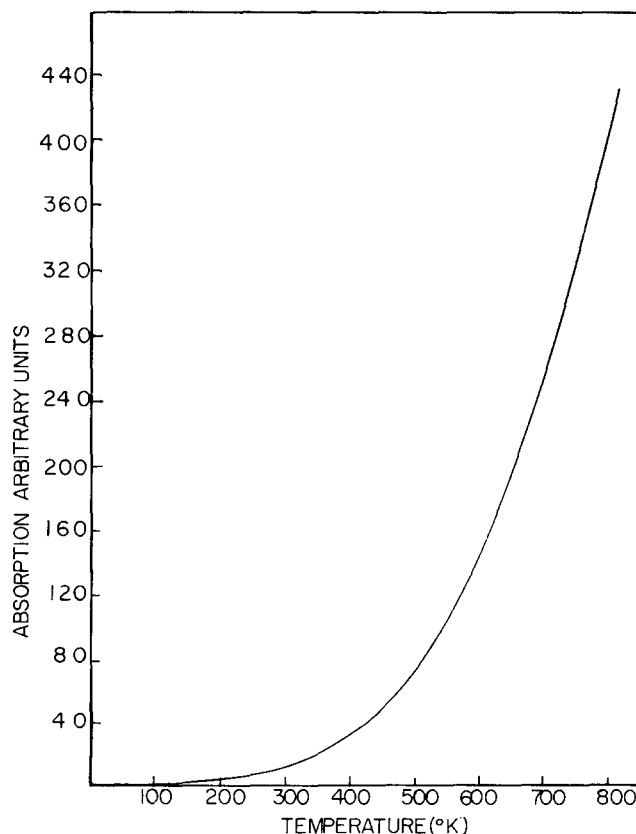


FIG. 1. Plot of the five-phonon intrinsic absorption at 10.6 μm as a function of temperature for KCl. (Note that the limiting value as $T \rightarrow 0$ is unity.)

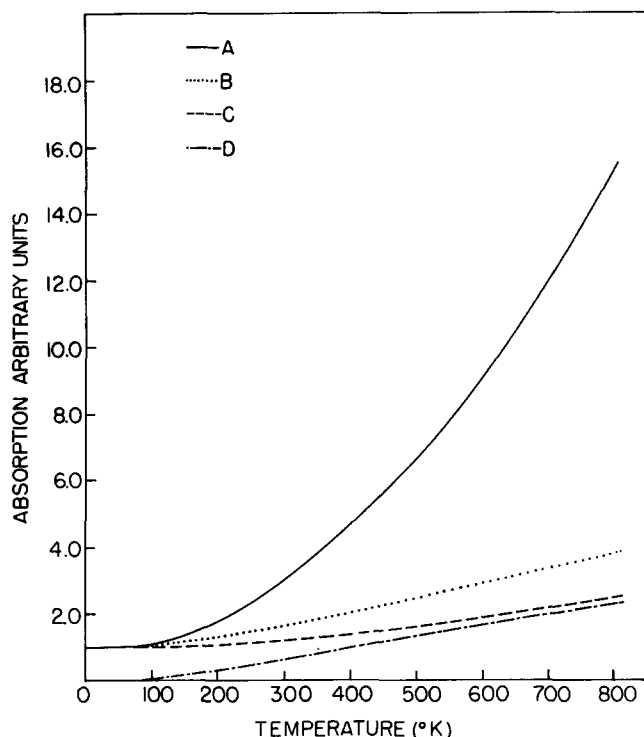


FIG. 2. Plots of various types of defect-induced absorption at 10.6 μm as functions of temperature for KCl: (A) absorption due to the creation of one local mode phonon and two lattice phonons; (B) absorption due to the creation of one local mode phonon and one band mode phonon; (C) absorption due to the first overtone of a local mode; (D) absorption due to the creation of a local mode phonon and the simultaneous destruction of a band mode phonon.

two-phonon side bands of the local mode absorption to occur at 10.6 μm . An alternative possibility is that the impurity mode frequency may be greater than 10.6 μm , and the absorption 10.6 μm is associated with a difference process in which a local mode phonon is created and a band mode phonon is destroyed. For the purposes of this letter we have chosen to examine four specific cases: (i) The absorption is due to the creation of a band mode phonon, whose frequency is taken to be the same as that used in the calculation of the intrinsic absorption, plus a local mode phonon. (ii) The absorption is due to the creation of a local mode phonon plus two band mode phonons. (iii) The absorption is due to the overtone of a local mode. (iv) The absorption is due to the creation of a local mode phonon together with the annihilation of a band mode phonon. The appropriate expressions for the temperature dependence of the absorption A are given by

$$A \propto (n_1 + 1)(n_0 + 1) - n_1 n_0,$$

$$n_1 = [\exp(\frac{4}{3}\hbar\Omega/kT) - 1]^{-1} \quad [\text{case(i)}]; \quad (2)$$

$$A \propto (n_2 + 1)(n_0 + 1)^2 - n_2 n_0^2, \quad (3)$$

$$n_2 = [\exp(\frac{2}{3}\hbar\Omega/kT) - 1]^{-1} \quad [\text{case(ii)}];$$

$$A \propto (n_3 + 1)^2 - n_3^2, \quad (4)$$

$$n_3 = [\exp(\hbar\Omega/2kT) - 1]^{-1} \quad [\text{case(iii)}];$$

$$A \propto n_0 - n_4, \quad (5)$$

$$n_4 = [\exp(6\hbar\Omega/5kT) - 1]^{-1} \quad [\text{case(iv)}].$$

These results are displayed graphically in Fig. 2.

On comparing the curves in Figs. 1 and 2, it is at once obvious that there is a great difference between the variation of absorption with temperature for the intrinsic five-phonon process and that predicted for processes (i)–(iv). In particular, the intrinsic absorption changes by more than an order of magnitude between 300 and 600 °K, while the defect-induced absorptions vary by factors of only 2 or 3 over the same range. The strongest variation is that manifested by the local mode plus two band mode phonons. However, this process is likely to be much weaker than the other possibilities since it is second order in the anharmonic interaction between the defect mode and the lattice modes while the other processes considered are only of first order in the anharmonicity.

Thus it would seem that measurements of the temperature dependence of the residual absorption could be an extremely useful diagnostic tool to discriminate between intrinsic absorption and impurity-induced absorption. For processes (i), (ii), and (iii) measurements from 200 °K upwards are likely to be the most revealing, for process (iv) measurements of the absorption at temperatures below 300 °K are likely to provide the most information, since the intrinsic absorption approaches a constant value below about 200 °K whereas the impurity absorption process (iv) decreases very rapidly with temperature below room temperature and becomes zero at 0 °K.

Thus we have demonstrated, by choice of certain specific examples, that measurements of the temperature dependence of the residual 10.6 μm absorption are a potentially powerful means of determining its origin. Moreover, more sophisticated theoretical studies are also likely to prove fruitful.

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