TRIPLE TRANSITIONS IN SOLID HYDROGEN AND DEUTERIUM

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The infrared absorption spectrum of the solid hydrogens is well understood in terms of a pairwise multipolar induction mechanism leading to the infrared activity of single and double transitions. However, in recent experiments we have identified various triple transitions for the first time\(^a\).\(^b\).

There are two basic mechanisms for the simultaneous excitation of three hydrogen molecules upon absorption of one photon of radiation:

- Intensity transfer due to mixing of states.
- Three-body induced dipole moments.

The infrared activity of triple transitions due to intensity transfer resulting from mixing of states was suggested by Tipping et al.\(^c\) As examples of this mechanism we present the transitions $Q_1(0) + Q_1(0) + S_0(0)$ and $S_1(0) + Q_1(0) + S_0(0)$ in both solid hydrogen and deuterium.

However, with this concept it is not possible to explain the triple transition $S_1(0) + Q_1(0) + Q_2(0)$ which we have also observed in both isotopic species of the hydrogen crystal. This transition is located at 12 788 cm\(^{-1}\) in solid para-H\(_2\) and at 9123.5 cm\(^{-1}\) in solid ortho-D\(_2\).

We present a theory of three-body induced dipole moments in order to account for the linestrength of this very remarkable transition. Experiments to observe the corresponding triple transition in solid HD are currently in preparation.