PROBING EXCITON DYNAMICS IN THE FREQUENCY DOMAIN

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We report high-resolution infrared absorption measurements of H_2 rovibrational transitions in chemically doped solid parahydrogen crystals which access both bound and delocalized exciton states. The excitons in this case correspond to vibrational (vibron) and rotational (roton) excitation of the H_2 molecules that make up the solid. Originally studied in the seminal work of Van Kranendonk,[a] our group and others are now studying how the exciton dynamics in solid parahydrogen are altered when the solid is doped with small concentrations of atoms. Recently, we have studied rare gas atoms and halogen atoms doped in solid parahydrogen. In the case of the halogen atom studies (Cl and Br to date), exciton decay channels include both reactive and non-reactive pathways. Special emphasis will be given to spectroscopic examples where the high-resolution lineshape reveals broadening and splitting due to delocalization of the exciton around the atomic impurity. Some of the lineshapes are understood in terms of the exciton dynamics they encode; however, some of the lineshapes await theoretical interpretation.

[a] J. Van Kranendonk, Solid Hydrogen: Theory of the Properties of Solid H_2, HD, and D_2 (Plenum Press, New York, 1983).