

PURE ROTATIONAL SPECTROSCOPY OF RARE GAS DIMERS BASED ON ROTATIONAL WAVE PACKET IMAGING

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We report time-domain rotational spectroscopy of argon dimer and krypton dimer by implementing time-resolved Coulomb explosion imaging of rotational wave packets. The rotational wave packets are created in the dimers with a ultrashort laser pulse, and their spatiotemporal evolution is fully characterized by measuring angular distribution of the fragment ions. The pump-probe measurements have been carried out up to a delay time of 16 ns. The alignment parameters, derived from the observed images, exhibit periodic oscillation lasting for more than 15 ns. Pure rotational spectrum of Ar₂ is obtained by Fourier transformation of the time traces of the alignment parameters. The frequency resolution in the spectrum is about 90 MHz, the highest ever achieved for Ar₂. The rotational constant and the centrifugal distortion constant are determined with much improved precision than the previous experimental results: $B_0 = 1.72713(9)$ GHz and $D_0 = 0.0310(5)$ MHz. The present B_0 value does not match within the quoted experimental uncertainty with that from the VUV spectroscopy, so far accepted as an experimental reference to assess theories. Spectrum of the krypton dimer will be also reported.