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TWO-PHOTON ABSORPTION OF ALKALI HALIDES USING SYNCHROTRON RADIATION

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Two-photon spectroscopy is very important and complementary to one-photon spectroscopy in the study of atoms, molecules and crystals, since the selection rules are very different and different sets of final states can be probed. Two-photon spectroscopy is a second order and, consequently, a very weak process. Its probability increases with the intensity of the two radiation sources, so that, at least one of them must be a laser. For this reason, two-photon spectroscopy has been limited to the long wavelength region, due to the lack of a suitable light source in the vacuum ultraviolet. As a matter of fact, in the case of alkali halides, two-photon absorption experiments have been carried out only for the alkali iodides and bromides[1], and the structure around the fundamental absorption threshold has been clarified.

We have overcome these limitations by using for the first time synchrotron radiation as a tunable light source, and a Nd-Yag laser in a two-photon experiment on KI, KCl, and NaCl. The experiment was performed on the vacuum ultraviolet beam line of the Italian synchrotron radiation facility PULS located at the Frascati National Laboratories of the INFN. The monochromatized ultraviolet radiation from the Adone storage ring crosses the alkali halide samples (cooled at 20 K) and then it is detected by a 56AVP photomultiplier through a sodium salicylate window. The laser beam crosses the samples at 90° with respect to the Adone beam. Particular care was used to minimize the laser radiation scattered towards the ultraviolet detector.

Since the Adone beam comes in pulses approximately 4 ns of duration and separated by 117 ns (8.53 MHz repetition rate), we had to synchronize each laser pulse (15 ns duration - 30 Hz repetition rate) with one of the Adone pulses. The train of pulses from the detector was processed with a two-channel boxcar integrator gated in such a way that one channel

averaged the pulses in coincidence with the laser and the other channel one of the pulses emitted by the same electron bunch orbiting in the storage ring, but out of coincidence. From the percentage difference between the two channels we derived the two-photon absorption coefficient β normalized to the laser intensity.

Fig. 1 shows the spectra of β for KI, KCl and NaCl as a function of the sum of the energies of the two-photons (laser + Adone). The two-photon spectra are compared with the one-photon absorption spectra found in the literature[2-4].

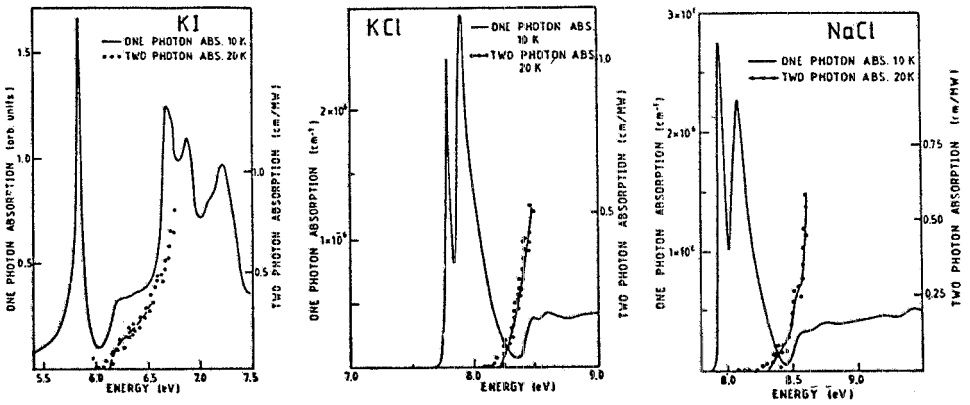

Fig. 1.

Our KI spectrum agrees well with the two-photon absorption spectrum of Frölich[1] measured up to 6.5 eV with the same type of laser and a conventional light source. The two-photon spectra of KCl and NaCl are very similar to that of KI, but their onsets at about 8.2 eV and 8.3 eV, respectively, do not occur in correspondence to the steps which are often attributed to the 2s excitons[3,4], but rather in the tail of the 1s exciton doublets. This is rather difficult to explain, because central cell corrections and electron-hole exchange are not expected to shift the 2p excitons from the 2s excitons by such a large amount ($\sim$0.2 eV).

The importance of the present results, that differ from previously reported laser+synchrotron radiation experiments[5,6] which actually were two-step absorptions, consists in having opened the vacuum ultraviolet region to such an important research tool as non linear optics. Several improvements to the first experimental apparatus are under development, such as doubling the Nd-Yag frequency, new ultraviolet detection systems, polarization analysis.

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