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STUDIES OF COMPOUND STATES OF NEGATIVE IONS USING LASER BEAMS

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R. N. Compton

Chemical Physics Section, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6125
and Department of Chemistry. The University of Tennessee Knoxville, Tennessee 37996

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I. Photodetachment Spectroscopy

This project began with our earlier studies of autodetachment spectroscopy of He^-, Be^-, and He_2^-. These experiments have been published in *Physical Review Letters* and summarized in two reviews. T. J. Kvale, a postdoctoral fellow from the University of Missouri, presented these studies in a recent invited talk at the Accelerator Conference (a review article was published, see publications). R. N. Compton also presented these results at the Gordon Research Conference on Photoelectron Spectroscopy; additionally, he presented this work in invited talks at the Pennsylvania State University and the University of Florida.

The studies of autodetachment of He_2^- into $He_2\,{}^1\Sigma_u^+$+e remains somewhat of a mystery. Professors Tim Heil (the University of Georgia) and R. J. Bartlett (the University of Florida) have worked with us on this problem. We find that our studies of autodetachmet spectroscopy of He_2^- giving ~15.7 eV electrons gives a considerably (~1 eV) lower potential energy function for $He_2\,{}^3\Sigma_u^+$ than presently accepted for this state. The theoretical analyses of Professors Heil and Bartlett indicate that autodetachment occurs from v=0,2,3,4, etc. of the $He_2^-\,{}^4\Pi_g$ state. Autodetachment from v=1 takes place leaving $He_2\,{}^3\Sigma_u^+$ in the v=0 state and a slow electron as we showed earlier. A paper combining both theory and experiment has been completed and will be submitted to *Physical Review* (see preprints).

One year ago we began studies of photodetachment spectroscopy of fast negative ion beams. The method has the advantage of producing ultrahigh resolution photoelectron spectra (<10 meV) in the center-of-mass system. Using this new apparatus we have obtained accurate electron affinities for B^- (boron minus) $He^-\,{}^4P_{5/2,3/2,1/2}$ and Ca^- (calcium minus). The Ca^- was shown to be bound with the configuration $Ca^-[Ar]4s^24p$. This is the first report of a bound state for a group IIA metal atom. Our studies prompted a theoretical calculation by Dr. C. Froese-Fischer of Vanderbilt University. A comparison theory–experimental paper was published in *Physical Review Letters*. These results have been well received by the atomic and molecular physics community and we have prepared a general write-up for the *New Scientist* (at their request). A similar, but more technical, write-up is being sent to *Physics Today*. These write-ups are included in the apprendix. Clearly, the results for Ca^- are the highlight of our studies for the year.

We have begun photoelectron angular distributions for negative ions. Figure 1 shows preliminary data for the angular distributions for photodetachment of $He^-(1s2s2p)^4P$ into $He(1s2s)^3S + e$. These results will be analyzed theoretically in conjunction with Dr. Hari Saha (the University of Florida).

The laser photodetachment studies described above were performed with a fixed frequency laser. In the coming year we plan to tune the dye laser and study negative ion resonances in the photodetachment continuum. Since we have electron energy analysis it is also possible to observe the resonances decaying into various possible channels. In addition, threshold measurements will allow us to obtain much higher accuracy and precision on electron affinity values and permit the determination of fine-structure energies for certain ions, e.g.

$$h\nu + Ca^{-}\,{}^2P_{1/2,3/2} \rightarrow Ca^{0}\,{}^3P_{0,1,2} + e. \quad (1)$$

In this particular case, four possible thresholds can be observed which will yield the fine-structure splitting for $Ca^{-}\,{}^2P_{1/2,3/2}$.

All of the measurements for B^-, He^-, and Ca^- were performed at power densities of $\sim 10^5$ W/cm^2 with only modestly focused laser beams. Plans for future studies include focusing the laser beam to reach power densities of $10^8 - 10^9$ W/cm^2 in order to examine multiphoton detachment and above-threshold detachment from a negative ion.

For completeness we include photoelectron spectra for B^- and He^- in Figs. 2 and 3, respectively. Electron affinity values were obtained for both of these atoms. Note that the electron spectra are obtained in the laboratory system. Conversion to the center-of-mass yields high resolution electron spectra.

All of these studies were performed at the Oak Ridge National Laboratory (ORNL). A photograph of the "crew" with the negative ion test facility is enclosed. Reading left to right — Tom Kvale (lab coat), David Pegg (beard), Jeff Thompson, (the kid), Bob Compton, and Gerald Alton.

II. Electron Scattering in the Laser Field

These experiments are being carried out both at the University of Tennessee (UT) and ORNL. The principal investigators are H. S. Carman, Jr., a new postdoctral fellow from Rice University, C. S. Feigerle, a new physical chemistry professor from the Joint Institute for Laboratory Astrophysics, the University of Colorado, and R. N. Compton, ORNL/UT. We have moved a vacuum chamber and experimental apparatus from ORNL to

UT and purchased a number of items with the grant from the Office of Naval Research. In particular, a new computer data acquisition set up was purchased and set-up by Feigerle. Only Dr. Carman's salary is being paid from the Navy grant. A third-year graduate student, John Manone, will be joining the project in January of 1988. We have ~80% use of the new departmental Nd:YAG pumped dye laser and wavelength extension system at our disposal. This apparatus is now assembled and will soon be used to study electron alkali atom collisions in the laser field.

The experiments at ORNL have been twofold. (1) Studies of above-threshold ionization (ATI) of alkali atoms, rare gas atoms, and nitric oxide and (2) collisions of ultraslow electrons with electron attaching molecules in the laser field. We will discuss each of these separately.

Above-threshold ionization represents a process in which a quasi-free electron adsorbs an extra electron in the continium following multiphoton ionization (MPI). Graduate students Adila Dodhy (University of Auburn) and Perry Blazewicz (Yale University) completed Ph.D. theses on this subject and their papers are included in the list of publications. Dr. Dodhy is now a postdoctoral appointee at the Max Planck Institute with Dr. H. Walther and T. Hansch and Dr. Blazewicz is a postdoctoral appointee in the Analytical Chemistry Division at ORNL. These studies are being carried on by Stanley J. Bajic, a graduate student in chemistry from UT.

Howard Carman has completed a study of six-photon MPI and ATI of nitric oxide. These results were reported at the Third International Laser Science Conference in Atlantic City. A long paper is being written in conjunction with Professor V. McKoy (California Institute of Technology). A short version of this work which will appear in the book of abstracts is included in the list of publications. Since Dr. Carman had not had previous experience with lasers. this project was designed to educate him in this area of future laser electron atom experiments.

Dr. Carman has begun a new set of experiments which are designed to study the reaction of high Rydberg state with electronegative molecules in the laser field. Since the Rydberg electron can be viewed as essentially "free" this can be considered as a means of studying electron scattering in the laser field. This is also analagous to the ATI studies described above. The experiment involves exciting cesium atoms to high Rydberg levels (n ~15-40) and crossing them with a supersonic nozzle beam of molecules. A high-power laser is also present as the exciting light or introduced separately (i.e., fundamental of the

YAG). Thus far we have examined the reaction

$$
\begin{aligned}
Cs^{*}(n = 10 - \infty) + SF_6 &\xrightarrow{h\nu} Cs^{+} + SF_6^{-} \\
&+ NO_2 \rightarrow Cs^{+} + NO_2^{-} \\
&+ (CH_3NO_2)_{n=1-5} \rightarrow\ nmCs^{+} + (CH_3NO_2)^{-}_{n=1-5}
\end{aligned}
\tag{2}
$$

Figure 4 shows a typical scan of SF_6^- signal versus laser wavelength under conditions of low laser power ($\sim 10^4$ W/cm^2).

Many new experimental conclusions are resulting from these studies and we will not go into them here. However, we will discuss an interesting effect which will illustrate the possibilities of this research.

Figure 5 shows a scan of the two-photon excitation of ns and nd states of cesium folowed by attachment to SF_6 molecules under conditions of high laser power ($\sim 10^8$ W/cm^2). We see that the nd levels are split into two peaks and the splitting increases with laser power. These results indicate that "quasi" free electrons are attaching to SF_6 molecules in the laser field. These results are being examined theoretically by Professor Peter Lambropoulos of the University of Southern California. We are encouraged by the possibilities of studying "quasi-free" ultraslow electron atom or molecule scattering in the laser field using high Rydberg atoms.

List of Figures

Fig. 1. Angular distributions for photodetached electrons from He^- (1s3s2p) via the process $h\nu + He^-(1s3s2p) \rightarrow He(1s2s)^3S + e$.

Fig. 2. Photodetachment electron spectra of Be^- ions. The ion beam energy was 40 keV and the laser wavelength was 594.4 nm.

Fig. 3. Photodetachment electron spectra of He^- ions. The ion beam energy was 40 keV and the laser wavelength was 599.4 nm.

Fig. 4. SF_6^- signal versus laser wavelength resulting from "attachment" of ultraslow Rydberg electrons (see Fig. 2). Both ns and nd Rydberg states are reacting with SF_6.

Fig. 5. SF_6^- signal at specific ns and nd levels under conditions of high laser power ($\sim 10^8$ W/cm^2). Note the symmetric splitting of the nd levels.

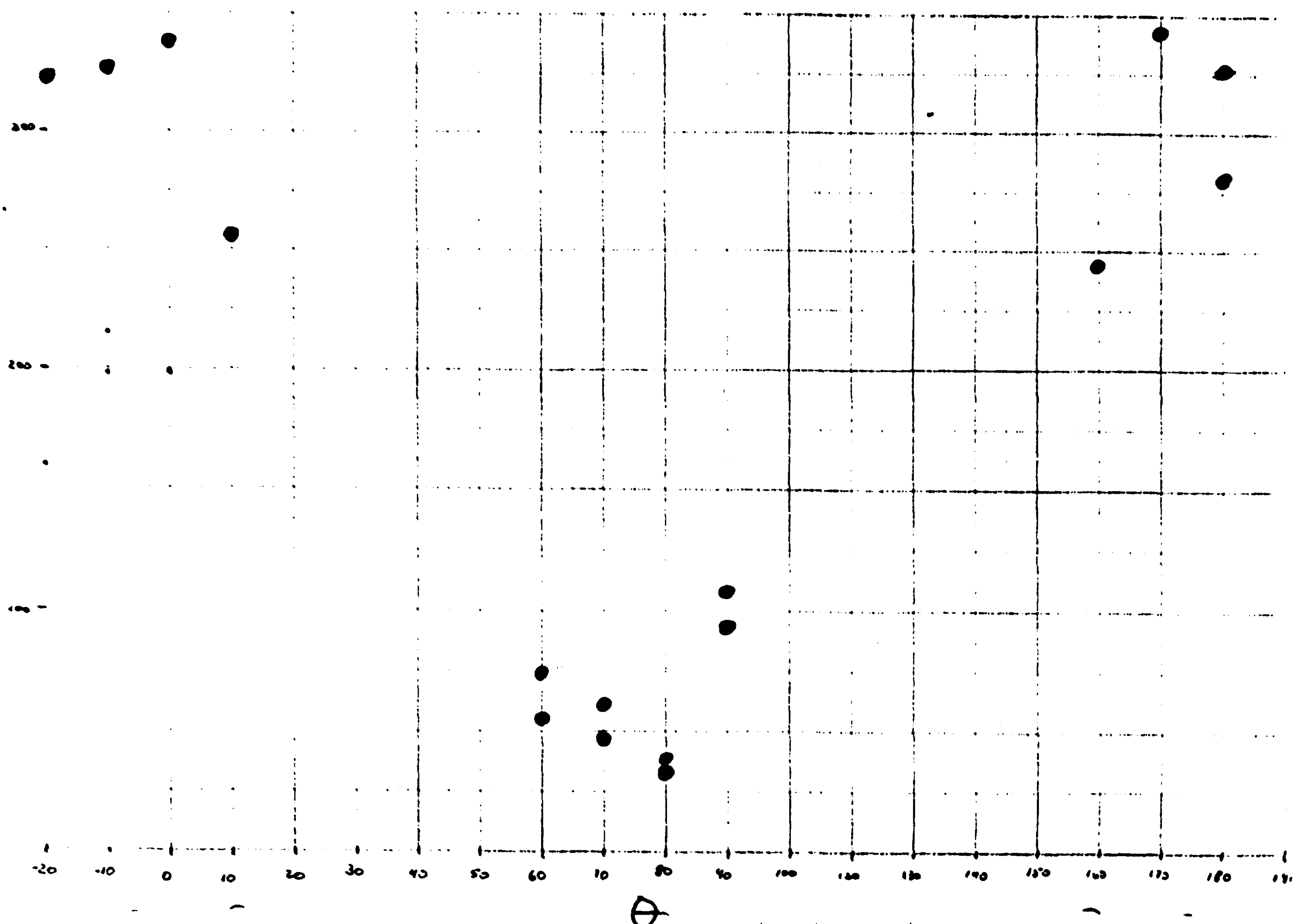
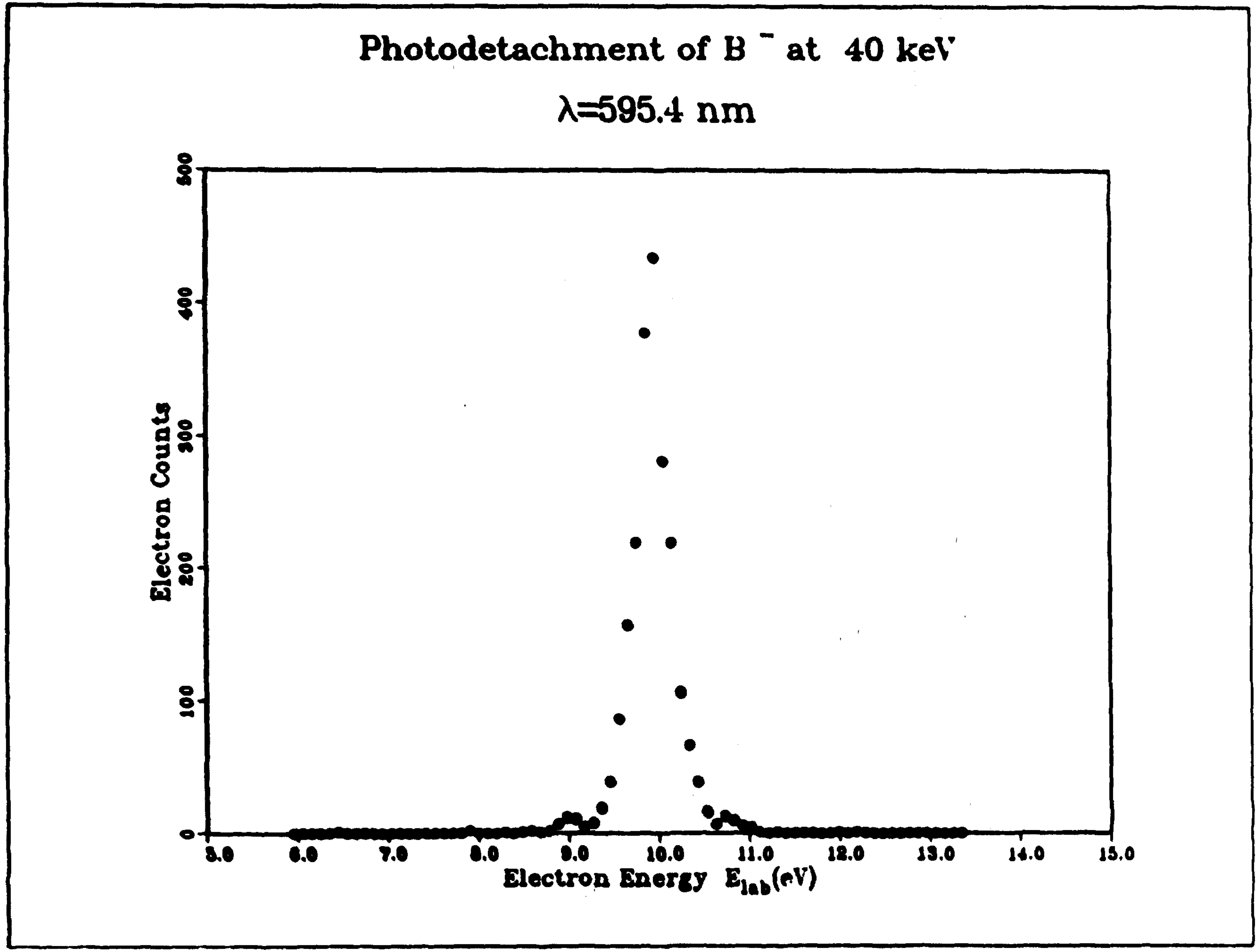
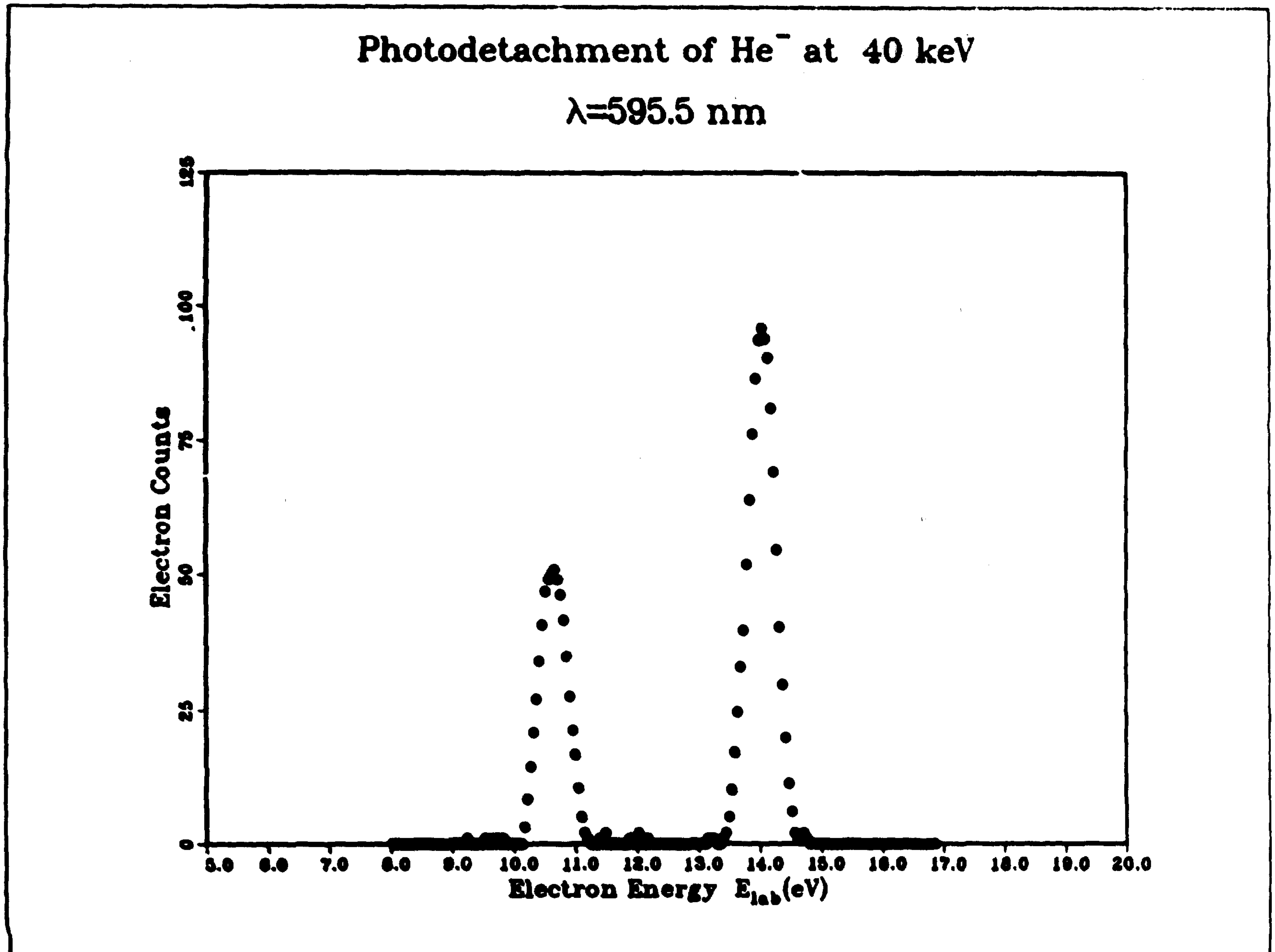


Fig 1



Photodetachment of B⁻ at 40 keV
λ=595.4 nm
Electron Counts
0
100
200
300
400
500
5.0
6.0
7.0
8.0
9.0
10.0
11.0
12.0
13.0
14.0
15.0
Electron Energy E_lab(eV)

Fig. 2



Photodetachment of He⁻ at 40 keV
λ=595.5 nm
Electron Counts
0
25
50
75
100
125
5.0 6.0 7.0 8.0 9.0 10.0 11.0 12.0 13.0 14.0 15.0 16.0 17.0 18.0 19.0 20.0
Electron Energy E_lab (eV)

Fig. 3

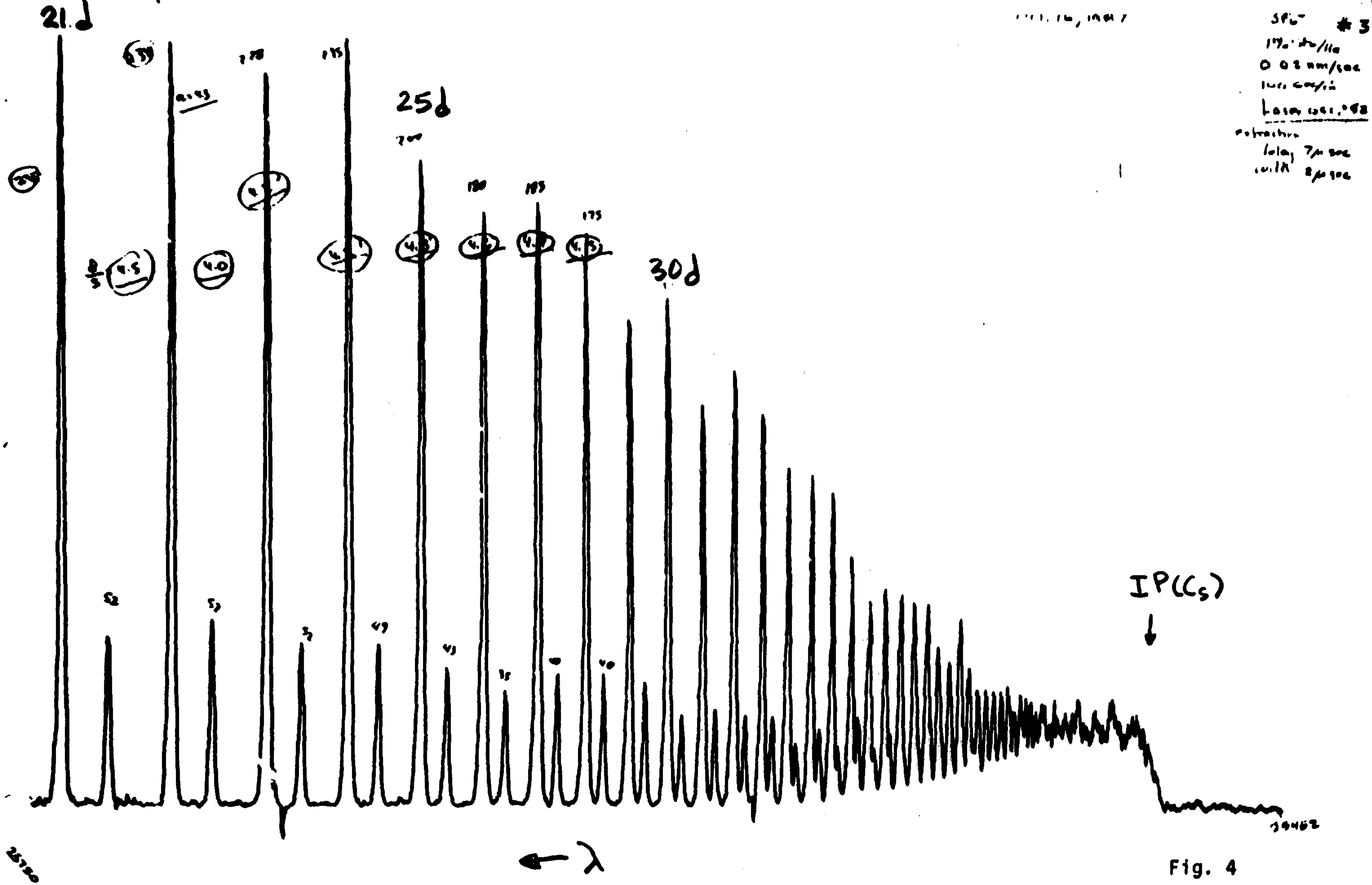


Fig. 4

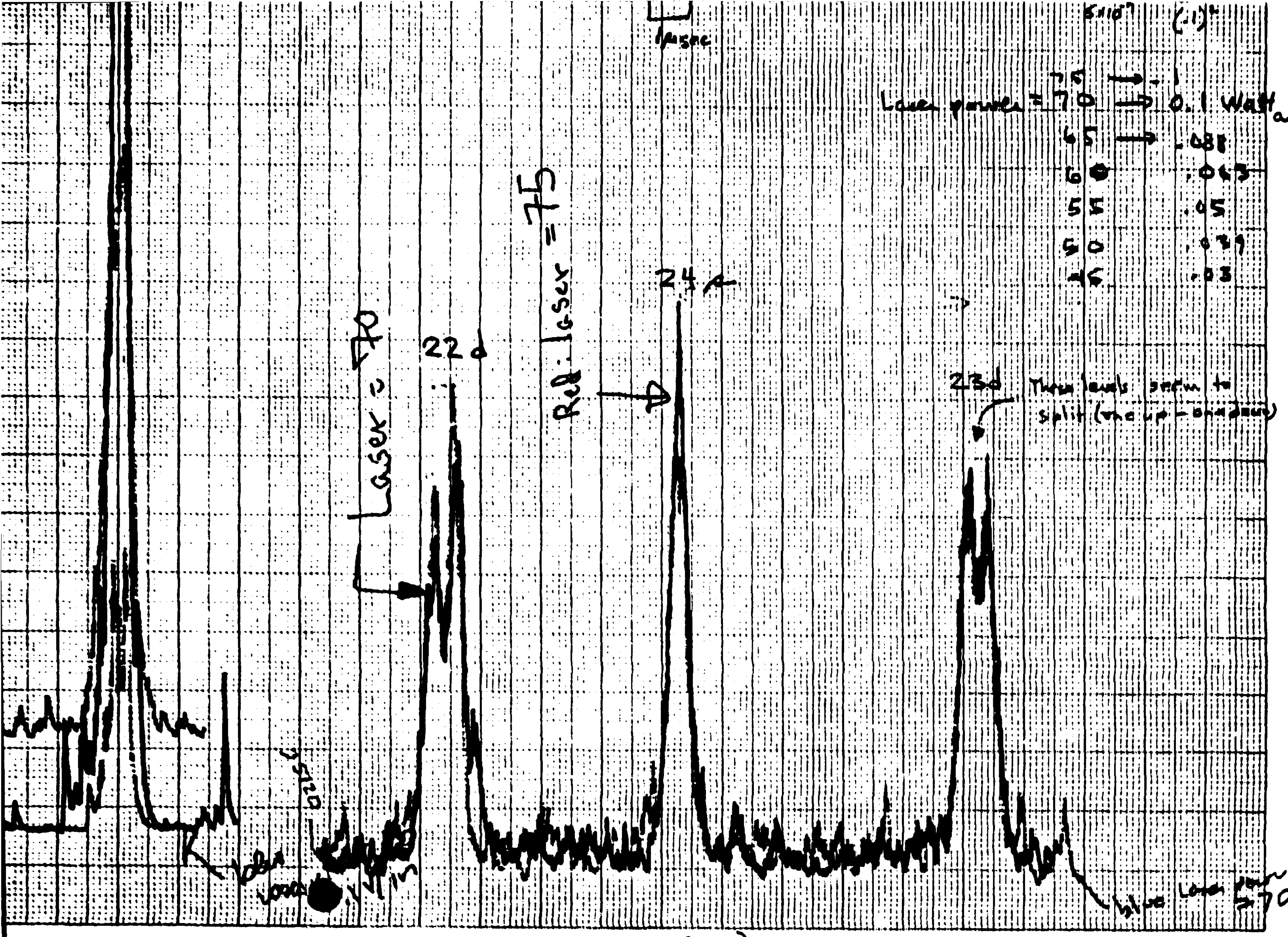

Laser power = 70 → 0.1 Watt ave
65 → .088
55 .05
50
45 .03
Laser = 70
22d
Ref. laser = 75
24
23d
These levels seem to split (one up - one down)
λ

Fig. 5