Title
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Publication Date
1988

LBL-24636

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Materials & Chemical Sciences Division

Presented at the Optoelectronic and Laser Applications in Science and Engineering Conference, Los Angeles, CA, January 10–17, 1988

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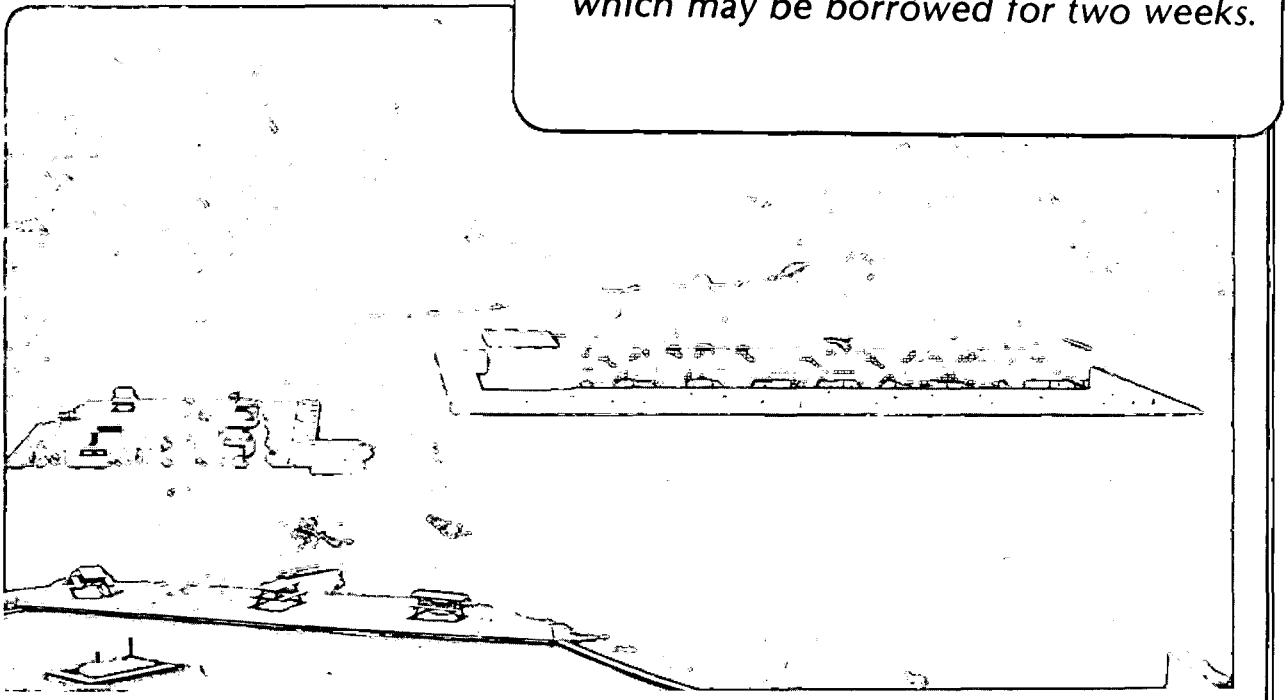
January 1988



Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

DISCLAIMER

XUV source for ultra-high resolution photoionization spectroscopy

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ABSTRACT

A tunable vuv-xuv laser system with good pulse energy (>100mJ in the visible) and narrow bandwidth (<210MHz in the xuv) was built in order to do ultra-high resolution photoionization spectroscopy. The capabilities of this system have been demonstrated with a study of isotope shifts and hyperfine splitting in Kr Rydberg levels.

1. INTRODUCTION

In the past years many photoionization studies of atoms, small molecules and clusters in a collimated beam have been done in our laboratory.[1] These were low resolution studies of the ionization threshold region. We are interested in studying such processes in more detail looking at individual transitions to Rydberg levels below and above the ionization threshold. Such experiments can be difficult for the following reasons. One, individual transitions to be observed potentially have very narrow linewidths because of the long lifetimes of Rydberg states in the absence of predissociation or autoionization. Two, above threshold photoionization cross sections are very small, 10^{-17} cm^2 to 10^{-20} cm^2, and large number densities for certain species (e.g. clusters) are hard to obtain, requiring an intense vuv source. Three, these studies involve using a wide range of photon energies making tunability over the broad vuv and extreme ultraviolet (xuv) regions a must.

We have chosen a laser source for these studies because of its versatility, spectral and temporal purity and good pulse energy. The laser system presented here is continuously tunable over much of the vuv and xuv regions. It has a high pulse energy of >100mJ in the visible and >30mJ in the ultra-

violet (uv) and a good beam quality. The bandwidth has been measured to be <210MHz ($0.007cm^{-1}$) in the xuv. The performance of this laser has enabled us to do experiments such as those mentioned above. One of these studies, that of Kr Rydberg levels, will be presented below.

2. DESIGN

There has been much effort in recent years to obtain ultra-narrow bandwidth laser light sources in the vuv and xuv.[2,3] In fact, many of the techniques used in our system have been employed previously. However, the way we have combined the components makes this system unique. The laser system is described in detail elsewhere[4] so only a brief discription will be given here. A schematic of the laser is shown in Figure 1.

The c.w. output of a Coherent 699-29 ring dye laser is pulse amplified by a dye amplifier chain pumped with the second harmonic of a Nd:YAG laser (Quantel model 592). The ring laser is optically isolated from the rest of the system by a permanent magnet Faraday isolator. This is necessary to prevent mode-hopping caused by the amplified fluorescence of the amplifiers. The dye amplifier is a three stage unit built in our laboratory. The dye cells are of the prism type first developed by D.S. Bethune[5] and have the dimensions 1mmu x 20mmlong, 3mmu x 30mm and 6mmu x 60mm. The 3mmu cell is double passed. These cells were used because they provide a uniform pumping and hence a uniform gain. There are also no problems with window damage because of the large pump volume and they are universal for different pump lasers.

The Nd:YAG laser is injection seeded for single-mode operation. This gives our amplified dye laser beam a smooth, nearly Gaussian, temporal profile. The injection seeded Nd:YAG laser also gives rise to a more stable uv and xuv power due to elimination of random modulations in the temporal profile of the pulse.

The pulsed visible beam is then frequency doubled to produce uv photons. The doubling is done in 25mm-30mm KDP crystals which are kept aligned while scanning by an auto-tracking unit (INRAD model 5-12 or Quanta-Ray model WEX1). To further extend our capabilities we can also mix the doubled light with the Nd:YAG fundamental to produce even shorter wavelength uv photons.

After generating the uv light we utilize four-wave mixing to produce radiation in the vuv and xuv spectral regions. This is done, as demonstrated previously,[6] by focusing the laser into a pulsed beam of Ar, Xe or CO. An improvement we have made in this technique involves the use of a large travel pulsed valve.[7] This new valve yields a higher number density in the mixing area compared to the source used previously. This, in turn, gives a conversion efficiency that is up to 100 times better than before.

A 1m normal incidence monochromator (McPherson model 225), used to seperate out the fundamental light beams, allows us to use the vuv and xuv photons without interference from the uv and/or visible beams and to recollimate the vuv-xuv beam allowing an optimal spatial overlap of the photon beam with our molecular beam. This is of significant importance in experiments where the signal is not abundant.

3. RESULTS

In this section the performance of the laser system will be discussed with regard to pulse energy, bandwidth, beam quality and tunability. Then some preliminary results of our Kr studies, showing the capabilities of the laser, will be presented. A more complete summary of the performance characteristics is given elsewhere.[4]

In the visible pulse energies of up to 120mJ have been measured. The output is above 100mj at the peak of some of the dyes used but falls off for the redder dyes. This is caused somewhat by the lower c.w. output in the red. A complete summary of the output energies for the different dyes used is

given in Figure 2. The beam is a 6mm circle with hard edges and some diffraction rings. We have been able to scan continuously from 562nm to 620nm using R590 in the 699-29 laser and various dyes in the amplifiers, again these are shown in Figure 2. A typical dye tuning range is about 10nm. The bandwidth has been measured to be ≈95MHz using a 300MHz etalon with a finesse of >75. The temporal profile is smooth but somewhat non-Gaussian.

Up to a 40% efficiency has been measured in converting the visible light to the uv. Typically, though, operating conversion efficiencies of 30-35% are realized. This gives useful uv energies of >30mJ at the peaks of the stronger dyes. The beam profile of the uv beam is very similar to that of the visible with the diffaction rings being more pronounced. When mixing the Nd:YAG laser fundamental with the doubled visible beam we have observed energies of >5mJ at 222nm. This process is limited somewhat by a mismatch of the two beam profiles giving poor overlap in the mixing crystal.

We have generated tunable light around wavelengths from 74nm to 124nm by using different mixing techniques. To test the resolution of our vuv source we scanned the 4P-7S $^2P_{3/2}$-(J=1) transition of Kr at 95.4nm. Under optimum conditions and correcting for Doppler broadening we found the xuv bandwidth to be <210MHz (see Figure 3).

This source has been developed to study the spectroscopy and dynamics of atomic and molecular systems in a collimated beam. The first concerted high-resolution study done with this system was of the 4P-nS $^2P_{3/2}$(J=1) (n=5,6,7) and 4P-nS $^2P_{1/2}$-(J=1) (n=5,6) transitions of Kr measured in a collimated beam at 123.6nm, 100.1nm, 94.5nm, 116.5nm and 95.1nm respectively.[8] In Figure 4 are shown the 7S $^2P_{3/2}$ transitions of the different Kr masses (M=78,80,82,83,84,86amu). Isotope shift measurements were make relative to the M=86amu transitions and the absolute measurements were made with a synchronously scanned 300MHz visible etalon calibrated with an I_2 cell. These were done for

all five transitions. The Kr isotopes are observed to exhibit a monotonic shift.

We have also observed the hyperfine splitting of Kr^{83} transitions. When an atomic nucleus has a non-zero nuclear angular momentum, I, there is coupling between this angular momentum and that of the electrons, J, given by $F=I+J, I+J-1, \ldots, |I-J|$. The only Kr isotope with a non-zero I is Kr^{83} with $I=9/2$. Accordingly, we observe three peaks for Kr^{83} corresponding to $F=11/2$, $9/2$ & $7/2$ and only one peak for the other masses in a given transition. The hyperfine splittings of Kr^{83} for the five different transitions studied are shown in Figure 5. The absolute values of the splittings were measured in the same manner as above. It is interesting to note that there are two trends observed. The first is a decrease in splitting with increasing n. This is expected because the further the excited electron is from the nucleus (the distance increases with n) the weaker the coupling will be. The second trend is the difference in splitting between the 1/2 and 3/2 series. A more detailed analysis of this work will appear in another publication.[8]

4. CONCLUSION

We are interested in ultra-high resolution studies of the photoionization processes of atoms, small molecules and clusters. In order to accomplish this we have developed a laser system with good vuv-xuv pulse energy, very narrow bandwidth and scanning capability throughout much of the vuv and xuv spectral regions. We have demonstrated the power of this laser with an indepth study of Kr Rydbery levels. Other experiments utilizing the capabilties of this laser are underway.

5. ACKNOWLEDGEMENT

This work is supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Chemical Sciences Division of the U.S. Department of Energy under contract no. DE-AC03-76SF00098. Part of the laser equipment used is contributed by the San Francisco Laser Center supported by the National Science Foundation under grant no. CHE83-03208.

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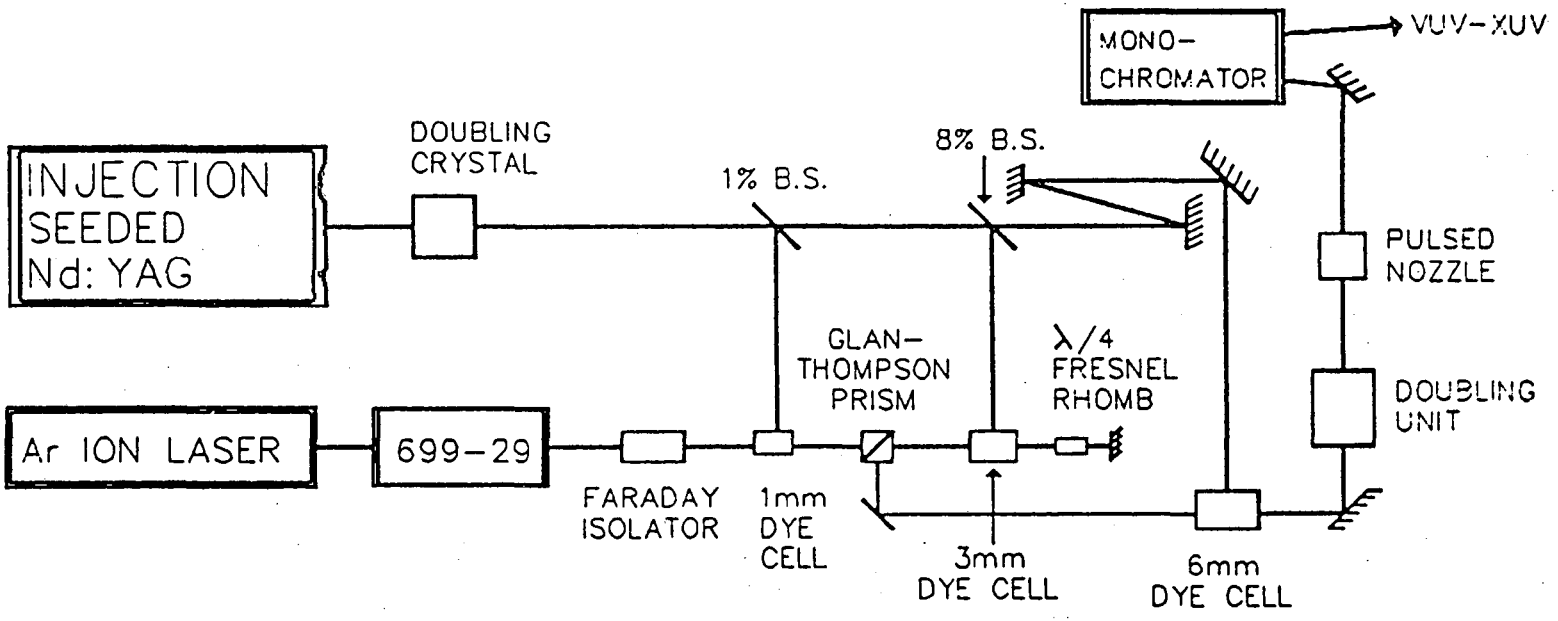

Figure 1. VUV-XUV laser system schematic.

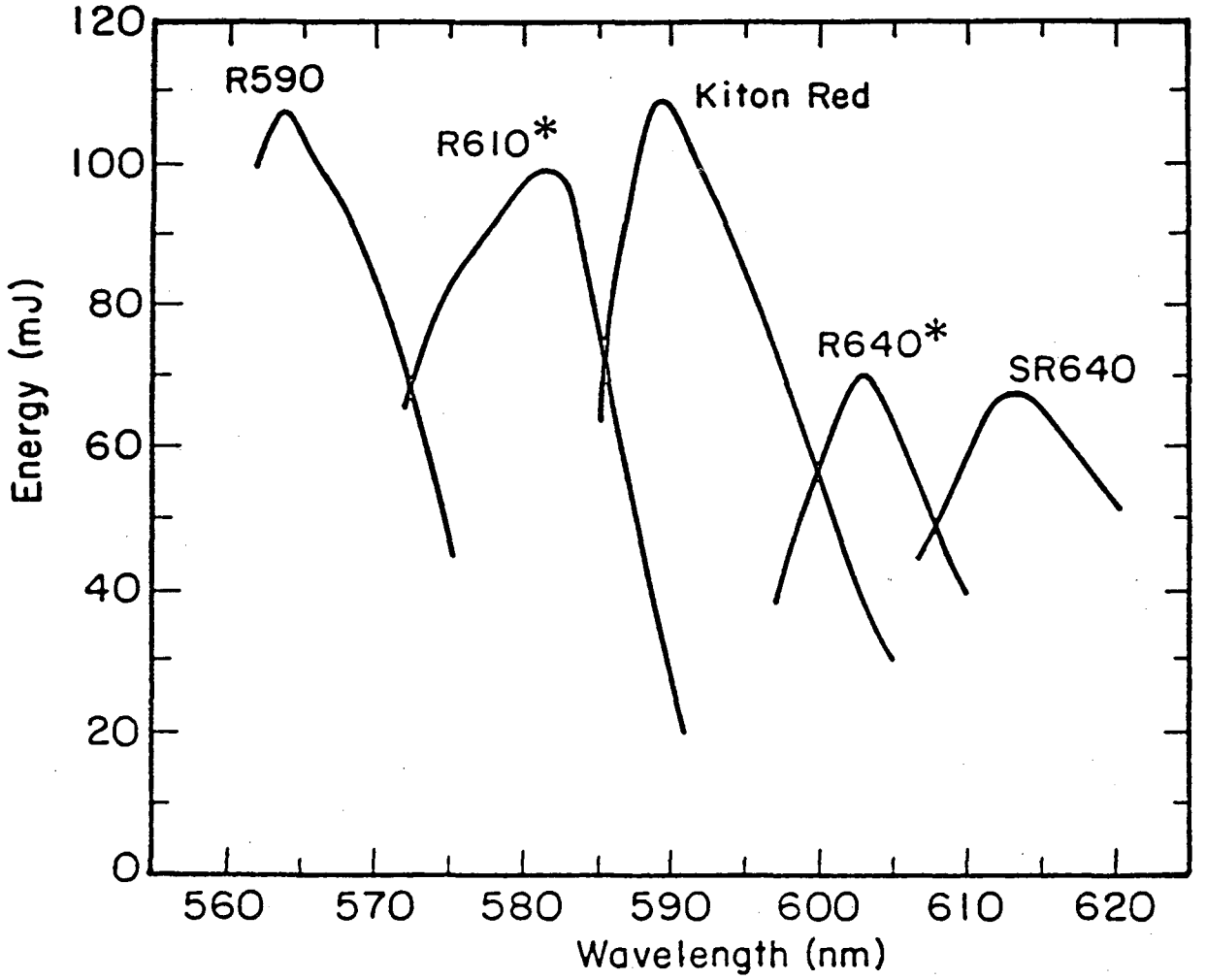

Figure 2. Laser output in the visible for different amplifier dyes (*) denotes basic solution.

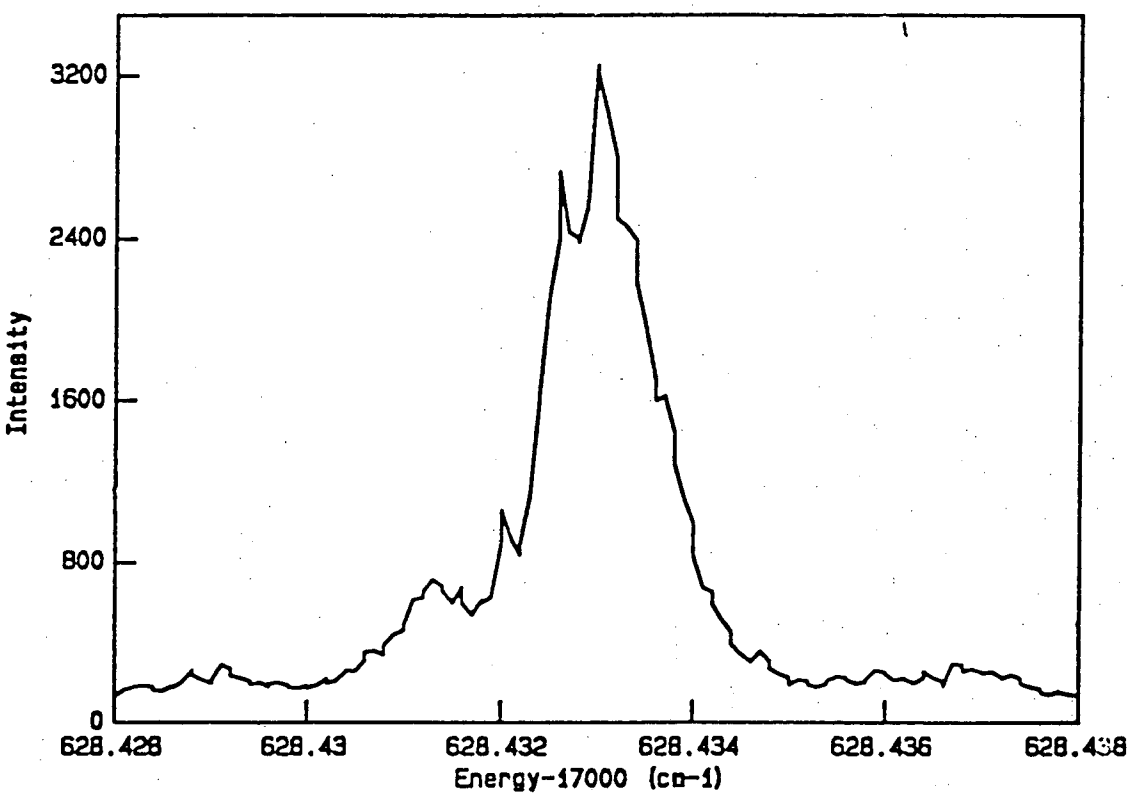

Figure 3. 4p-7s $^2P_{3/2}(J=1)$ transition for Kr^{86}. Energy units are for visible wavelength ($\nu_{xuv}=6\nu_{vis}$). $FWHM_{xuv}$ is $0.007cm^{-1}$.

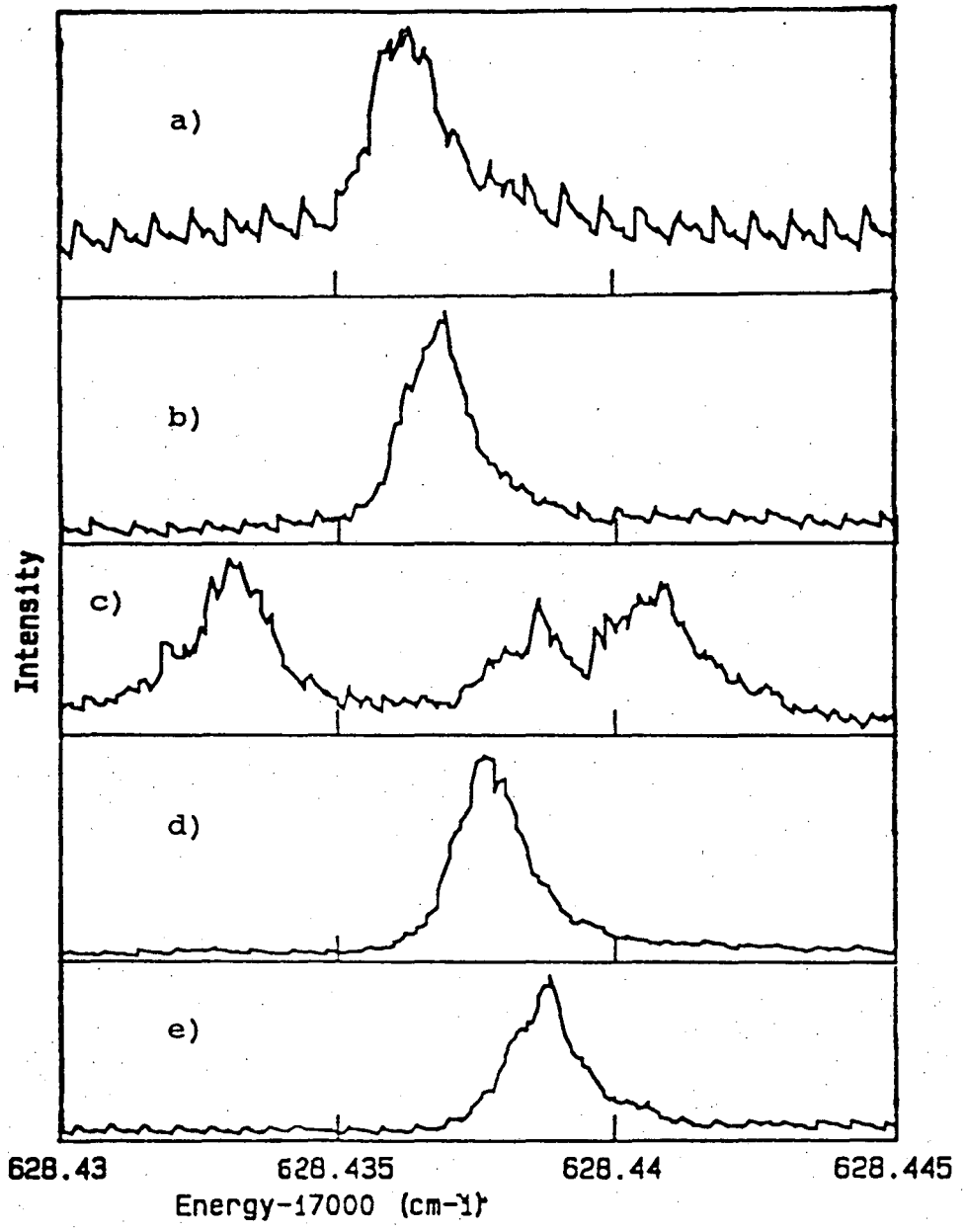

Figure 4. 4p-7s $^2P_{3/2}(J=1)$ transition for different Kr isotopes. Energy units are for visible wavelength ($\nu_{xuv}=6\nu_{vis}$). (a) Kr^{80}. (b) Kr^{82}. (c) Kr^{83}. (d) Kr^{84}. (e) Kr^{86}. (Kr^{78} is not shown.)

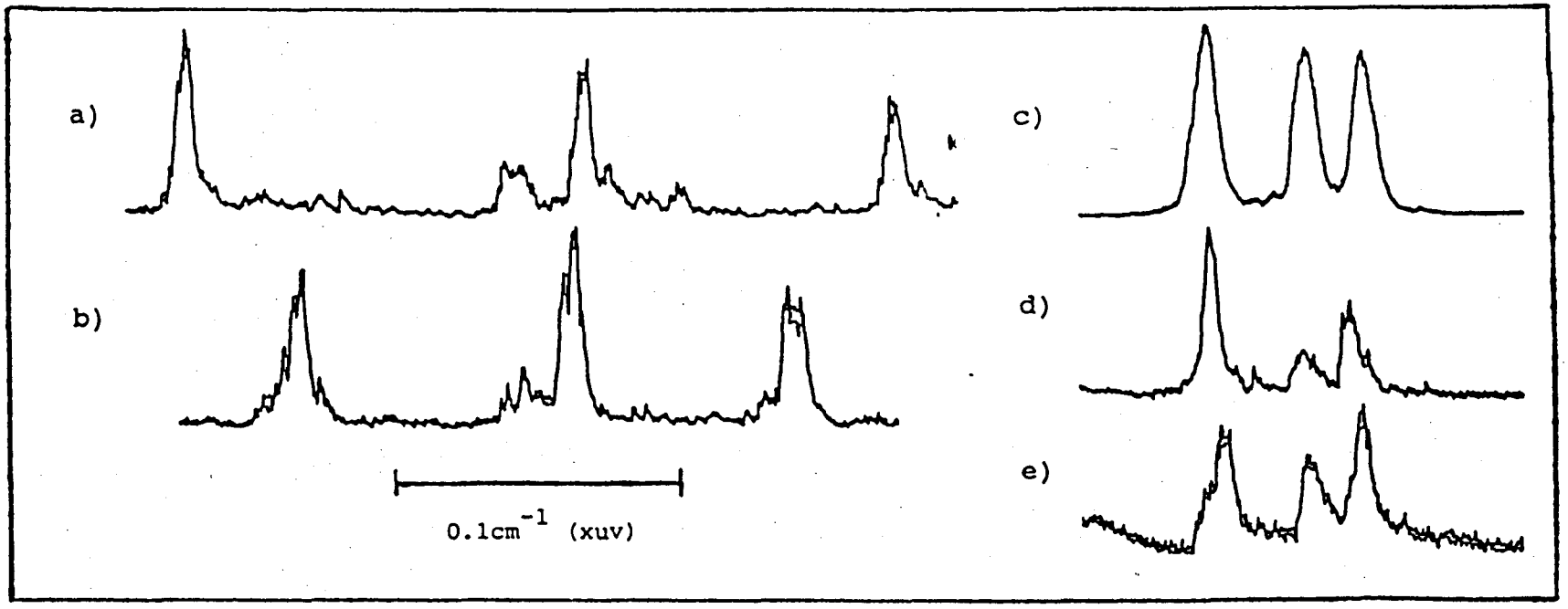

Figure 5. Hyperfine splitting of Kr^{83} for the five different transitions studied. a) 4p-5s $^2P_{1/2}$(J=1). b) 4p-6s $^2P_{1/2}$. c) 4p-5s $^2P_{3/2}$. d) 4p-6s $^2P_{3/2}$. e) 4p-7s $^2P_{3/2}$.

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