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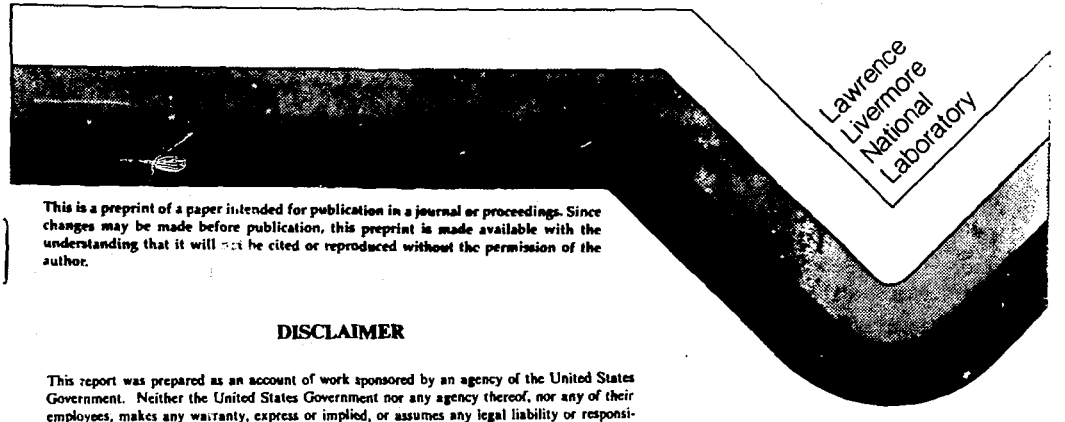

DISCLAIMER

CALCULATIONS OF PHOTOABSORPTION BY ATOMS USING A LINEAR RESPONSE METHOD

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ABSTRACT

We have made extensive calculations of photoabsorption by all neutral atoms from hydrogen to lawrencium for photon energies up to one kilovolt. Our method was the relativistic time-dependent local density approximation with the usual configuration average for open shells. The most important collective effects are included through an induced field. Expected features such as resonant photoemission and autoionization are seen. Examples of the calculations will be shown. The computer program used is available from the Computer Physics Communications Program Library.

Several years ago Liberman and Zangwill [1] developed a relativistic local density computer program for doing linear response calculations of photoabsorption in atoms. The program was designed (so far as possible) to be efficient and reliable. Taking advantage of idle time on the Los Alamos National Laboratory Cray computers, extensive calculations have been done on all atoms ($1 \leq Z \leq 103$) for photon energies up to 1 kilovolt. It is hoped that our results will be published eventually; our intention here is to advertise these unpublished results with a few examples and to point out the availability of the computer program [1] (and a companion non-relativistic program [2]) from the Computer Physics Communications Program Library.

A few words about the nature of the calculations [3]: In the first part a set of orbital functions are obtained as solutions of the Dirac equation,

$$(c\underline{\alpha}\cdot\underline{p} + \beta mc^2 + v^0(\underline{r}) - E_i)\,\psi_i^0(\underline{r}) = 0 \quad , \tag{1}$$

with a self-consistent field potential function,

$$v^0(\underline{r}) = -\frac{Ze^2}{r} + \int \frac{e^2\rho^0(\underline{r}')}{|\underline{r}-\underline{r}'|}\,d\underline{r}' + v_{xc}(\rho^0(\underline{r})) \quad , \tag{2}$$

where the charge density is obtained from the orbital functions,

$$\rho^0(\underline{r}) = \sum_i |\,\psi_i^0(\underline{r})\,|^2 \quad . \tag{3}$$

An iterative procedure is used to make E_i and ψ_i^0 consistent with $\rho^0(\underline{r})$ and $v^0(\underline{r})$. Equations (1), (2), and (3) determine the initial state of the atom. In the next few equations, the time-dependent orbital functions,

$$\psi_i^0(\underline{r},t) = \psi_i^0(\underline{r})e^{-iE_it/\hbar} \quad ,$$

are used.

The atom is exposed to a photon field, and first order time dependent perturbation theory is used to calculate the changes in the orbital functions. The perturbation to an orbital function is a solution to a time-dependent Dirac equation,

$$(c\underline{\alpha}\cdot\underline{p} + \beta mc^2 + v^0(\underline{r}) - i\hbar\frac{\partial}{\partial t})\psi_i^1(\underline{r},t) = -v^1(\underline{r},t)\,\psi_i^0(\underline{r},t) \quad , \tag{4}$$

where the perturbing field has two parts:

an external photon field,

$$v^1_{ext}(\underline{r},t) = \varepsilon\, z \cos \omega\, t, \tag{5a}$$

and an induced field,

$$v^1_{ind}(\underline{r},t) = \int \frac{e^2 \rho^1(\underline{r}',t)}{|\underline{r} - \underline{r}'|}\, d\,\underline{r}' + \rho^1(\underline{r},t)\, \frac{\partial}{\partial \rho^0}\, v_{xc}(\rho^0(\underline{r})). \tag{5b}$$

The induced charge density is computed from the orbital functions and their perturbations,

$$\rho^1(\underline{r},t) = \sum_i\ \psi^0_i(\underline{r},t)^*\, \psi^1_i(\underline{r},t) + c.c. \tag{6}$$

The solution of the inhomogeneous Dirac equation utilizes Green functions. Again, an iterative procedure is used to solve equations (4), (5), and (6). The photoabsorption cross sections are obtained from matrix elements of the perturbing potential. When only the external photon field is used,

$$\sigma(i\to j) \propto |\ (\psi^0_j \mid v^1_{ext} \mid \psi^0_i)\ |^2\ ; \tag{7}$$

we call this the independent particle approximation (IPA). When both external and induced fields are used,

$$\sigma(i\to j) \propto |\ (\psi^0_j \mid v^1_{ext} + v^1_{ind} \mid \psi^0_i)\ |^2\ ; \tag{8}$$

we call this the linear response approximation (LRA).

Our first example is the total photoabsorption cross section of manganese (Fig. 1). The two approximations -- IPA and LRA -- are compared with the data from Henke's [4] compilation. Henke uses both experimental and calculated cross sections in his compilation, and the agreement between our IPA calculation and his data is probably the result of his data for manganese coming from another IPA calculation. Some recent measurements [5] of manganese partial photo cross sections (Fig. 2) show that the linear response calculation is in fact the right one. Other measurements of photoabsorption by manganese have been made by Bruhn, et al [6, 7]. Calculations using many body perturbation theory by Garvin, et al [8] and using the random phase approximation by Amusia, et al [9] are similar to ours in most respects.

In the case of xenon (Fig. 3), agreement between Henke's data [4] and the LRA is better because Henke was able to draw on good experimental data.

Neither LRA nor IPA results agree very well with Henke's [4] uranium data (Figure 4) between 200 and 500 ev. Wendin and Del Grande [10] have shown that the discrepancy is due to poor experimental values in this energy region. The case of uranium is probably not unique. Our extensive calculations represent a valuable overall view of atomic photoabsorption, and in much of the vacuum ultraviolet region may be the best source for absorption cross section data.

A last example from our calculations concerns Cooper minima (Fig. 5 and 6). The matrix element $(3p|z|d)$ has a zero near $\hbar\omega = 40$ eV, and the corresponding IPA partial cross section also is zero at that energy. The LRA partial cross section has a minimum near 50 eV. A minimum was also found for the 3s→p LRA partial cross section at about the same energy but not for the IPA cross section. In trying to understand this, we observed that the 3s→p partial cross section has a Cooper minimum for elements with Z = 11, 12, 13, 14, and 15 according to both the IPA and LRA. For Z = 16, 17, and 18 the increasing threshold for this partial cross section overtakes the minimum according to the IPA, but in the LRA the minimum remains above threshold. Amusia, et al [11] noted this minimum in the argon partial cross section in 1972. They provide a rather different explanation from the one suggested here, but it should not be difficult to reconcile the two points of view.

Acknowledgments

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Figures Captions

Fig. 1. Total cross section for manganese according to IPA and LRA theories and Henke's compilation.

Fig. 2. Manganese 3d partial cross sections -- theory and the Oak Ridge experiments.

Fig. 3. Total cross section for xenon according to IPA and LRA theories and Henke's compilation.

Fig. 4. Total cross section for uranium according to IPA and LRA theories and Henke's compilation.

Fig. 5. Calculated partial cross sections (3p1/2 to d3/2) for argon from IPA and LRA theories.

Fig. 6. Calculated partial cross sections (3s1/2 to p1/2) for argon from IPA and LRA theories showing an induced minimum.

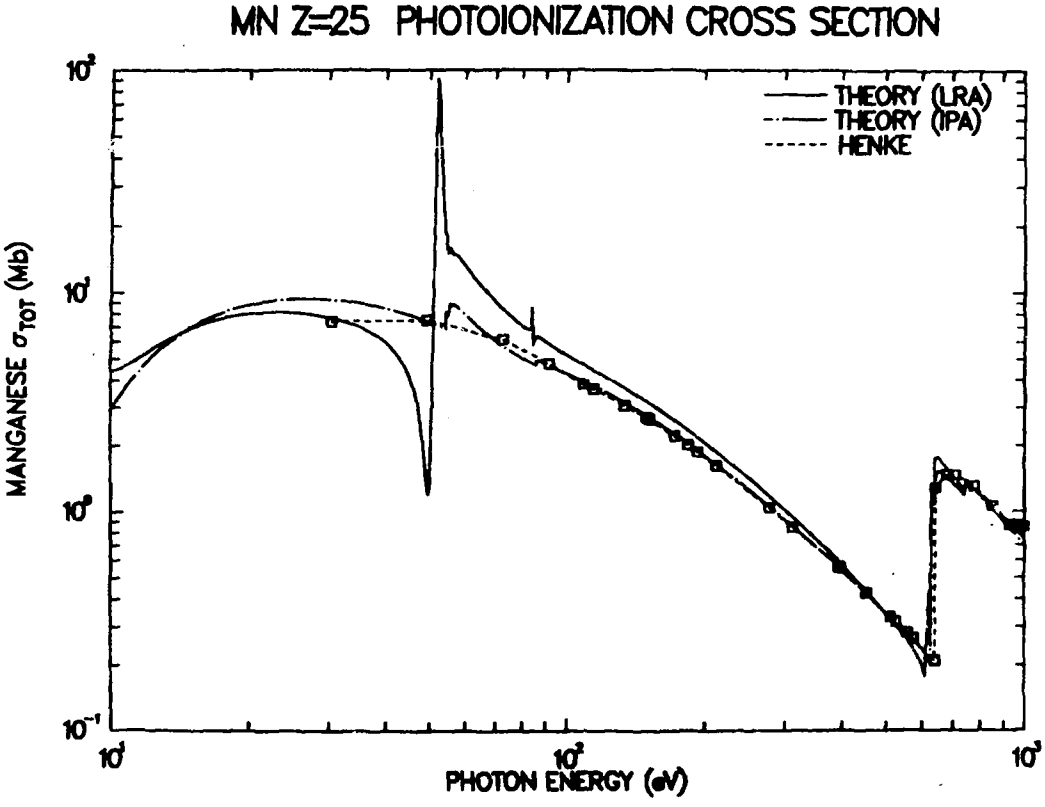
MN Z=25 PHOTOIONIZATION CROSS SECTION
THEORY (LRA)
THEORY (IPA)
HENKE
MANGANESE σ_{TOT} (Mb)
PHOTON ENERGY (eV)
10^2
10^1
10^0
10^{-1}
10^1
10^2
10^3

Figure 1.

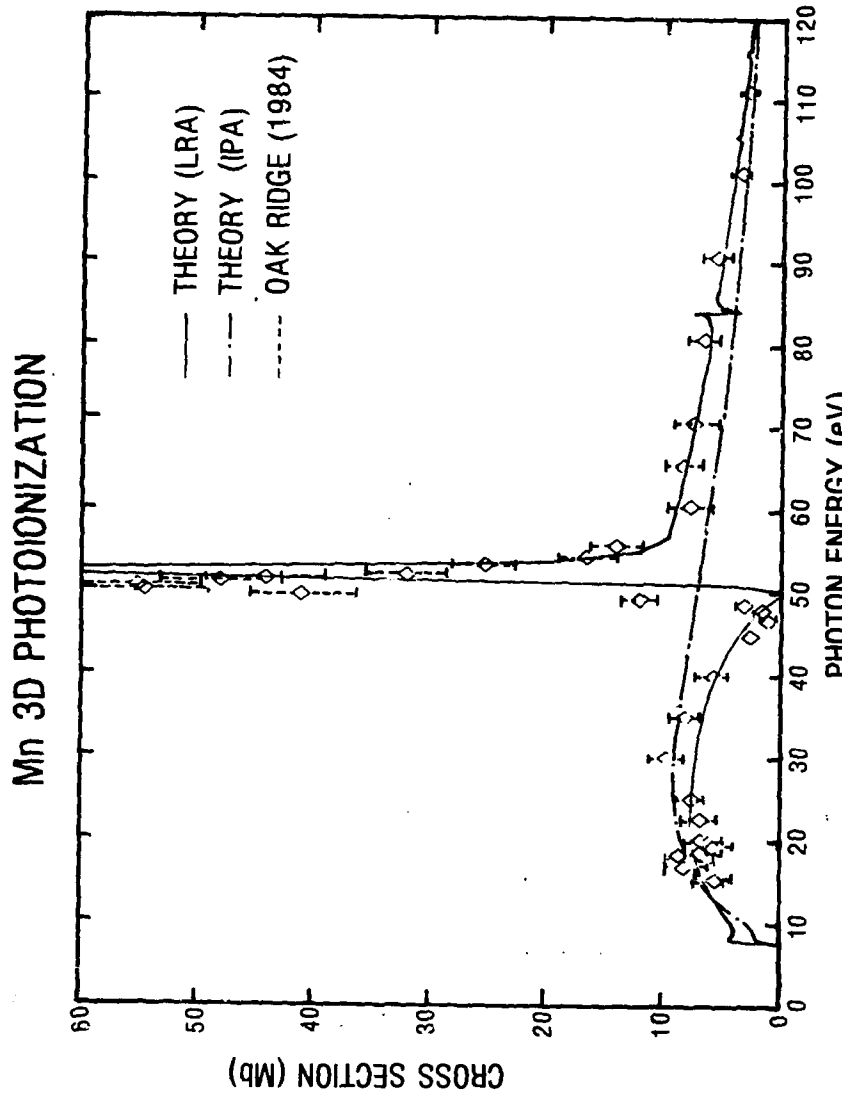
Mn 3D PHOTOIONIZATION
THEORY (LRA)
THEORY (IPA)
OAK RIDGE (1984)
CROSS SECTION (Mb)
PHOTON ENERGY (eV)
0
10
20
30
40
50
60
70
80
90
100
110
120

Figure 2

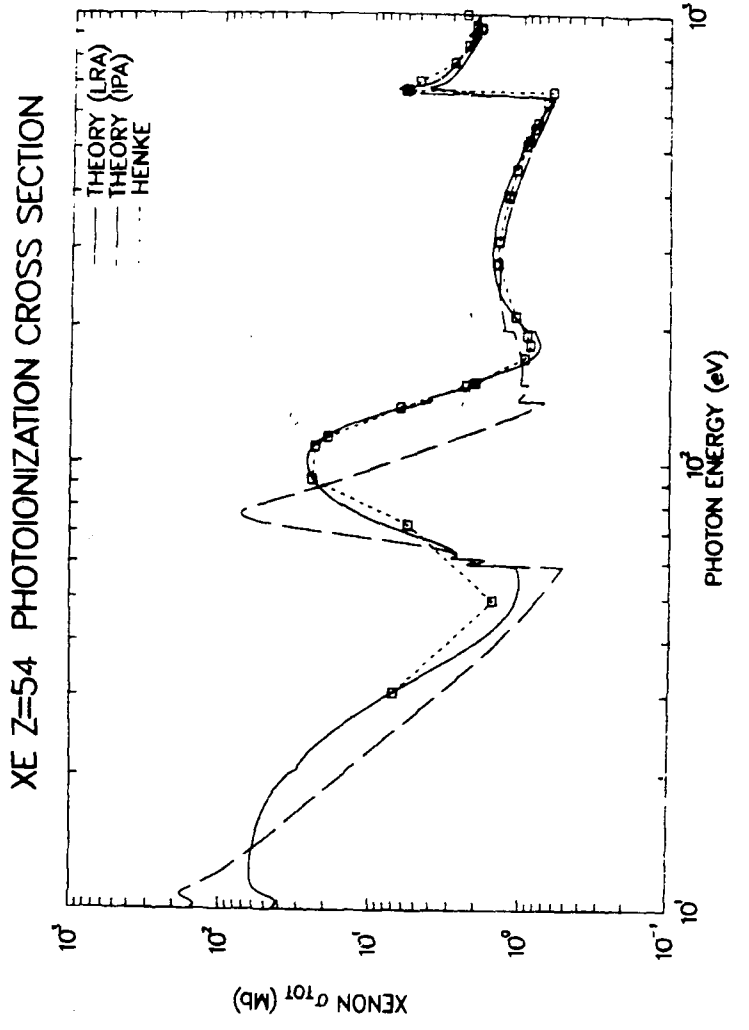
XE Z=54 PHOTOIONIZATION CROSS SECTION
THEORY (LRA)
THEORY (IPA)
HENKE
XENON σ_{TOT} (Mb)
PHOTON ENERGY (eV)
10^3
10^2
10^1
10^0
10^{-1}
10^1
10^2
10^3

Figure 3

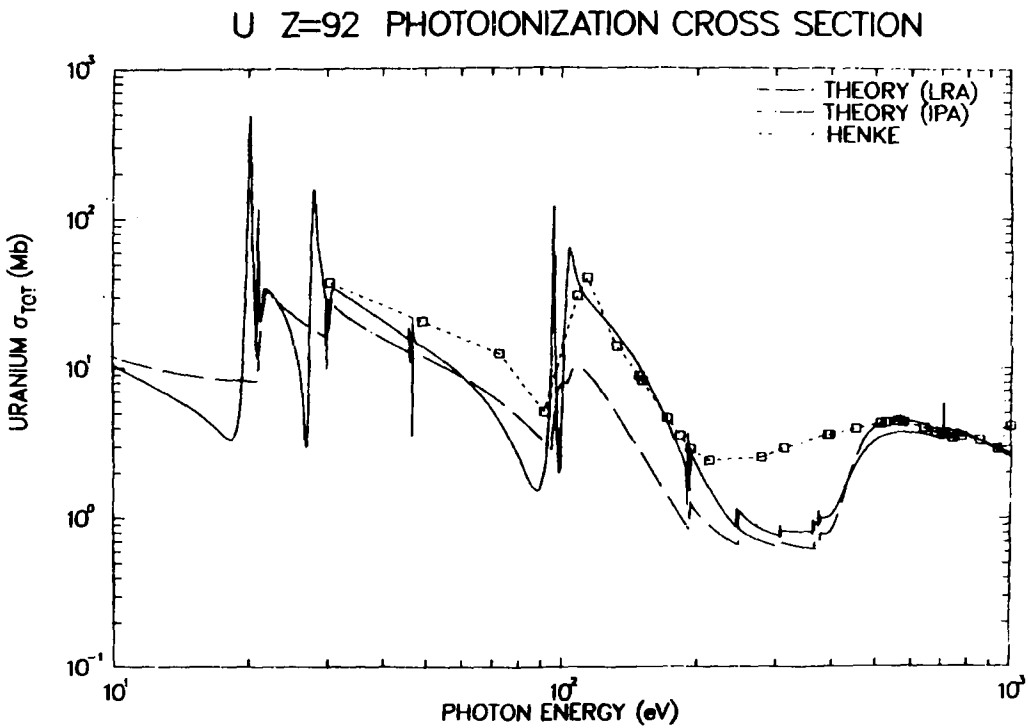
U Z=92 PHOTOIONIZATION CROSS SECTION
THEORY (LRA)
THEORY (IPA)
HENKE
URANIUM σ_{TOT} (Mb)
PHOTON ENERGY (eV)
10^3
10^2
10^1
10^0
10^{-1}
10^1
10^2
10^3

Figure 4

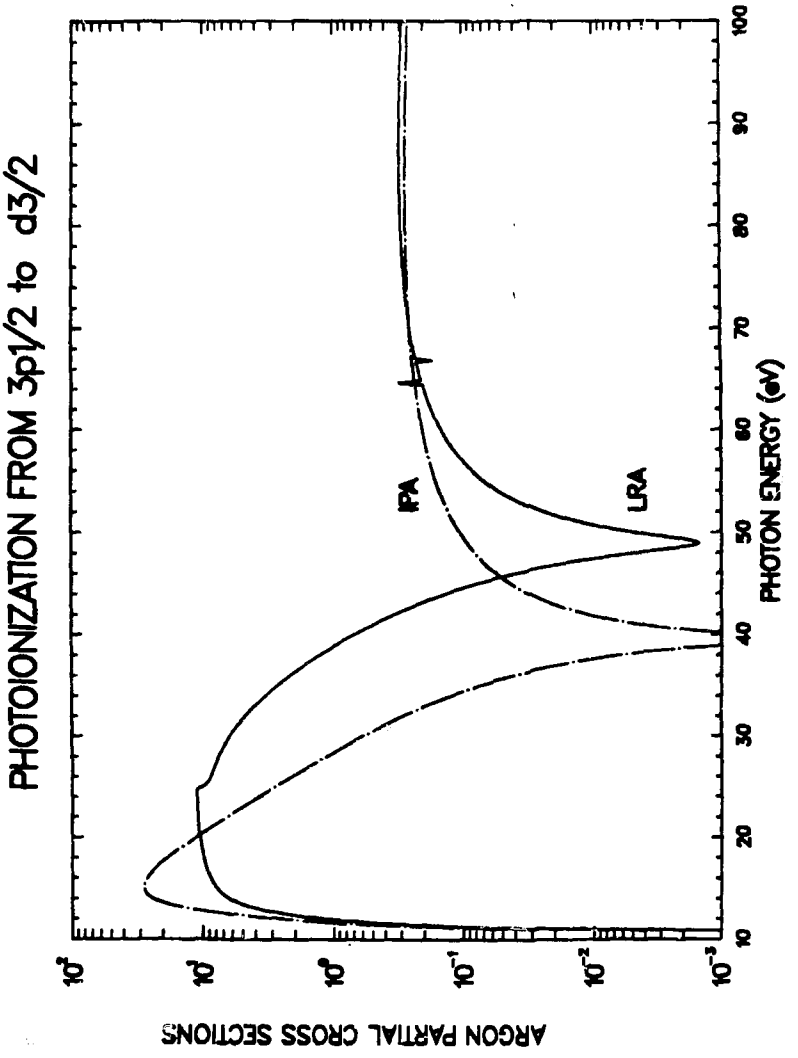
PHOTOIONIZATION FROM 3p1/2 to d3/2
IPA
LRA
ARGON PARTIAL CROSS SECTIONS
PHOTON ENERGY (eV)
10
20
30
40
50
60
70
80
90
100
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}

Figure 5

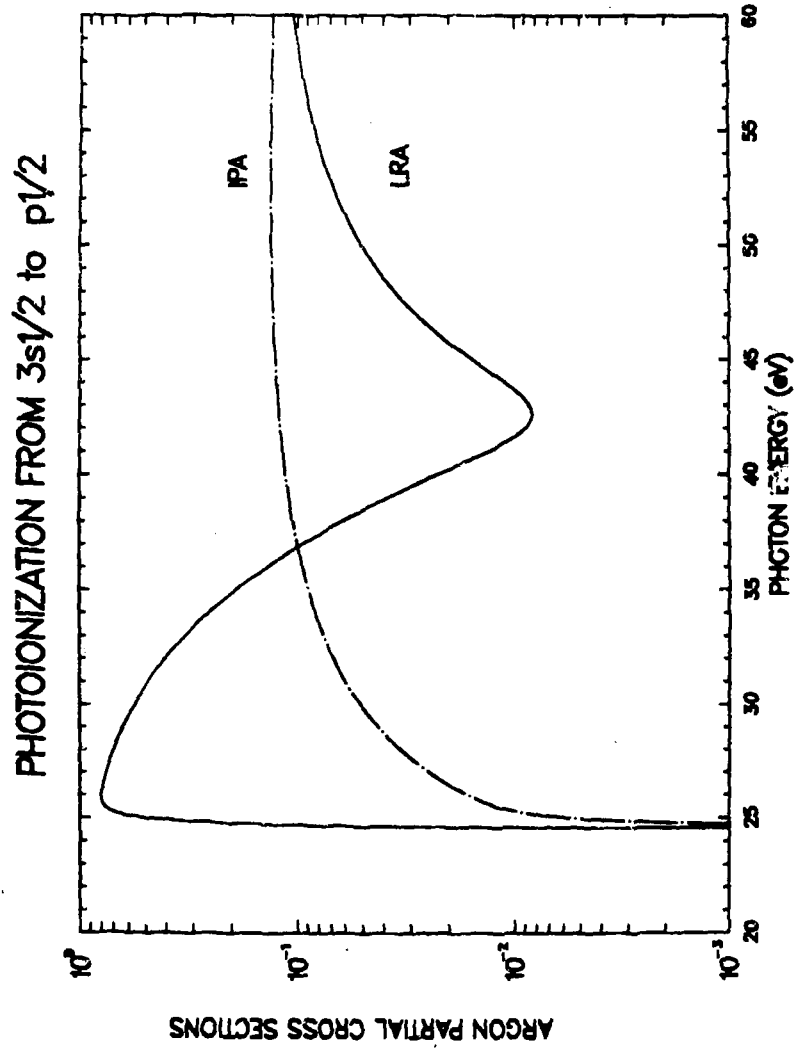
PHOTOIONIZATION FROM 3s1/2 to p1/2
IPA
LRA
ARGON PARTIAL CROSS SECTIONS
PHOTON ENERGY (eV)
10^0
10^-1
10^-2
10^-3
20
25
30
35
40
45
50
55
60

Figure 6