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EXPERIMENTAL EVALUATION OF NEW SOLID STATE LASER SCHEMES.(U)
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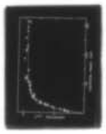

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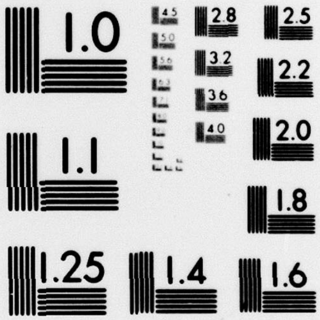

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Various schemes for tunable solid state lasers are discussed. Results of measurements of excited state absorption for YAG:Ce^{3+} are presented. The potential of color center lasers for blue-green emission is examined.

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The Final Report to the
Office of Naval Research
(Physics Section - Arlington)
For Contract No. N00014-76-C-0402
entitled

"Experimental Evaluation of New Solid State Laser Schemes"

by
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We report here the results of our investigation of new solid state laser schemes during the 1977-78 contract period. The emphasis has remained on identifying promising allowed transitions of ions or centers in crystalline solids since these are characterized by broad absorption and emission bands which facilitate pumping and permit tuning. For this final contract period the research has concentrated in two areas: Firstly, we have carefully investigated the excited state absorption (ESA) found in YAG:Ce^{3+} because of the major implications it has for all potential $4f^{n-1}5d \rightarrow 4f^{n}$ laser schemes. Secondly, we have examined F-center complexes in alkali halides as another possible source of blue-green stimulated emission.

As part of our survey we examined YLF:Ce^{3+}, another likely candidate for a tunable laser. Pump bands were found at 250 nm and 286 nm; at these wavelengths there are no simple and inexpensive pumps which can provide sufficient excitation. The emission peaks in the ultraviolet at 325 nm, well outside the acceptable limits for a blue-green laser. For these reasons we discontinued work on this system although subsequently other investigators obtained stimulated emission from YLF:Ce^{3+}.[1]

We have made a detailed study of the transient absorption first observed in this laboratory for YAG:Ce^{3+} with the objectives of identifying its origin, determining its magnitude, and measuring its spectrum.[2,3] This transient absorption consists of a part which decays rapidly (called the short component) and a part which decays more slowly (long component). Our investigation shows conclusively that the short component is Ce^{3+} ESA with the lowest 5d band as the initial state; the long component is most likely due to absorption by excited centers in the

YAG host.

A block diagram of the apparatus is shown in Figure 1. The Ce^{3+} ions were excited by pumping the 334 nm absorption band with a nitrogen laser and monitoring the attenuation of a probe beam passing through the excited region (Figure 2).

Our experiments show clearly that the short component is associated with the Ce^{3+} ion: it is present in all Ce^{3+} doped samples regardless of origin or concentration, but it is absent in all undoped samples. The lifetime of the short component (68 ns) is the same for all samples and is identical to that of the lowest Ce^{3+} 5d level.[2] This makes it unlikely that the initial state for this process is an upper 5d band (lifetime ≤ 2 ns[4]). These observations were later verified by other investigators.[5]

We have additionally measured the ESA spectrum over most of the visible spectrum and find indications of a peak in the red or near infrared (Figure 3). This peak is at the expected position for a $5d \rightarrow 5d$ transition and eliminates the possibility that an unrelaxed ^2F level is the initial state for the ESA. The short component ESA was measured as a function of excitation intensity and was fitted with a theoretical curve (Figure 4).[3] The observed saturation (due to every Ce^{3+} ion within the probed volume being excited) not only provides additional evidence that the absorption is due to Ce^{3+} but also allows us to obtain an accurate value for the ESA cross section, $\sigma^* = 1.4 \times 10^{-19}$ cm^2 at 633 nm. Additionally, we find the magnitude of the ESA to be unchanged at 77 K.

This cross section is much larger than would be expected for the parity-forbidden 5d→5d transition. One possible explanation is the admixture of opposity parity states from higher lying configurations into the 5d levels. Another is that the transition is from a 5d state to either a charge transfer band or an opposite parity configuration whose levels have been strongly split by the local ligand field.

In contrast, the long component is found to be extremely sample dependent both with respect to magnitude and lifetime. It is also observed in undoped YAG samples although for these the lifetimes are three orders of magnitude shorter and the absorption is a factor of five weaker. In addition, the long component lifetimes are nonexponential, suggesting a recombination process often associated with centers in crystals. Detailed results will be presented in reference 3, but our conclusion is that the long component of ESA is associated with ultraviolet excitation of defects already present in the YAG host. Some of these centers may be associated with Ce^{3+} ions or may be involved in an energy transfer process with the Ce^{3+}. In any case, losses due to the long component at realistic pump levels are not as significant as those due to the Ce^{3+} ESA and can probably be reduced through advances in materials preparation.

Our investigation of YAG:Ce^{3+} as a model system for tunable rare earth lasers has produced significant results with regard to the development of $4f^{n-1}5d \rightarrow 4f^{n}$ lasers in general and the blue-green laser program in particular. The most important implication is that one cannot <u>a priori</u> neglect ESA losses from transitions which would be forbidden for an isolated ion. Since both the wavelength of $4f^{n-1}5d \rightarrow 4f^{n}$ transitions and the existence of ESA are host-dependent, our investigation indicates that one must examine potential systems on a case by case basis.

In addition to rare earth lasers we examined the feasibility of color center lasers for visible operation. Several complexes of alkali halide F-centers have been use for highly efficient lasers in the infrared.[6,7] We investigated the possibility of extending these results into the visible spectrum using F_2 and F_3^+ centers in LiF since these lie closest to the wavelength range of interest. In order to obtain experience with these systems we built and operated a tunable color center laser using F_2^+ centers in LiF.

A number of major problems were found, the most serious of which is the ease with which these centers are bleached. Although oscillation has been reported at 670 nm for the F_2 center in LiF,[7] this center is quickly photo-ionized through a two-step process even at low pump levels[8] and as a lasing material would probably have a useful lifetime of only seconds. The F_3^+ center (emission maximum at 525 nm) is equally sensitive to two-step bleaching.

There are, in addition, materials problems associated with these centers. It is impossible to produce crystals with only one type of center present and the absorption bands of undesired centers generally overlap those of the desired center. This makes effective pumping impossible unless the undesired centers are decolored by a selective process. Many of these centers are unstable at room temperature and to maintain a sufficiently high concentration it is necessary to either keep the crystals permanently at reduced temperature or to develop techniques to grow then with impurities which are effective room temperature electron traps.

In conclusion, we feel that at least for the present alkali halide color centers hold little promise as blue-green laser materials. Allowed transitions of rare earths in solids still have great potential but must be individually evaluated for unforeseen loss mechanisms.

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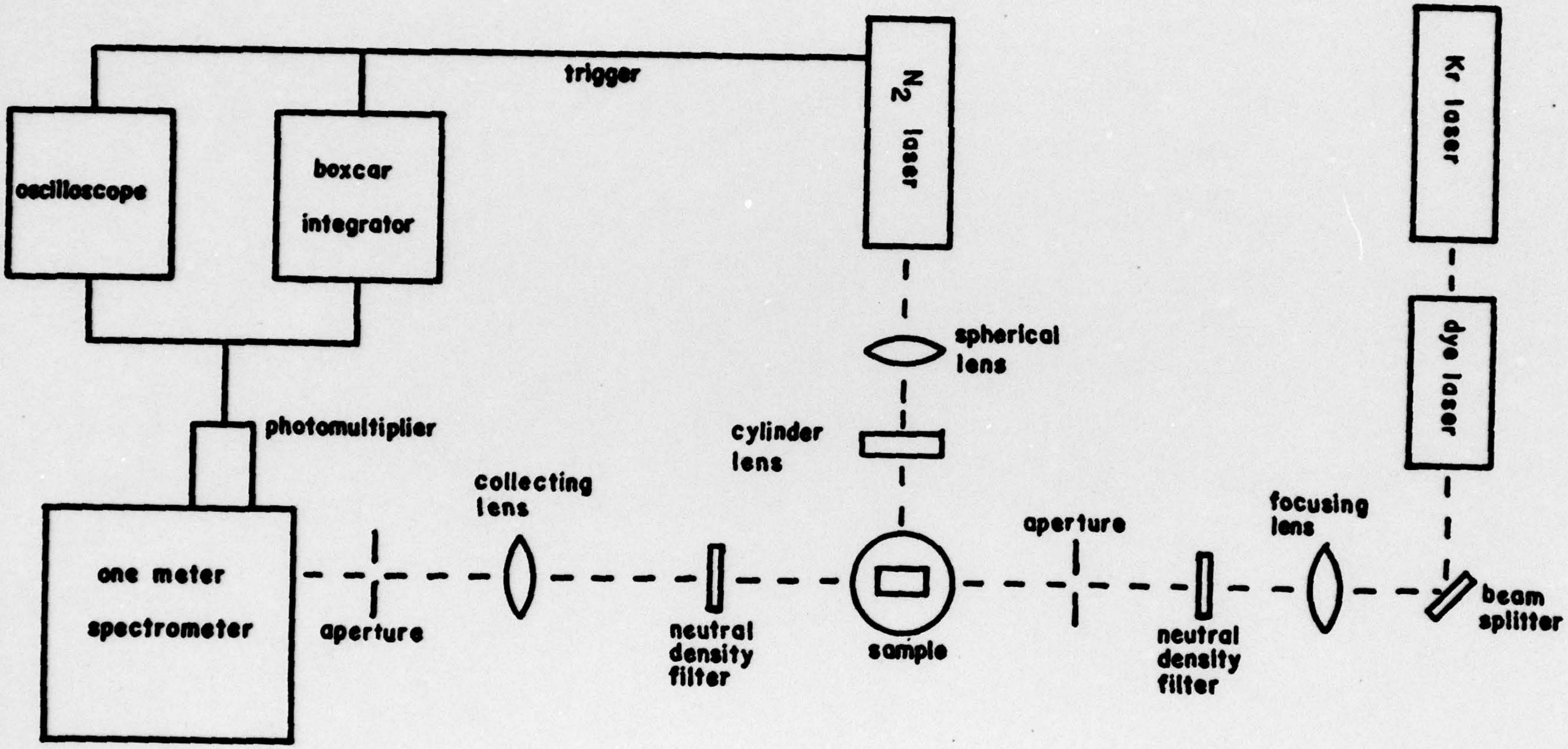
Figure 1. Block diagram of apparatus for measuring ESA.

Figure 2. Sample geometry for ESA measurements showing overlap between probe beam and excitation beam.

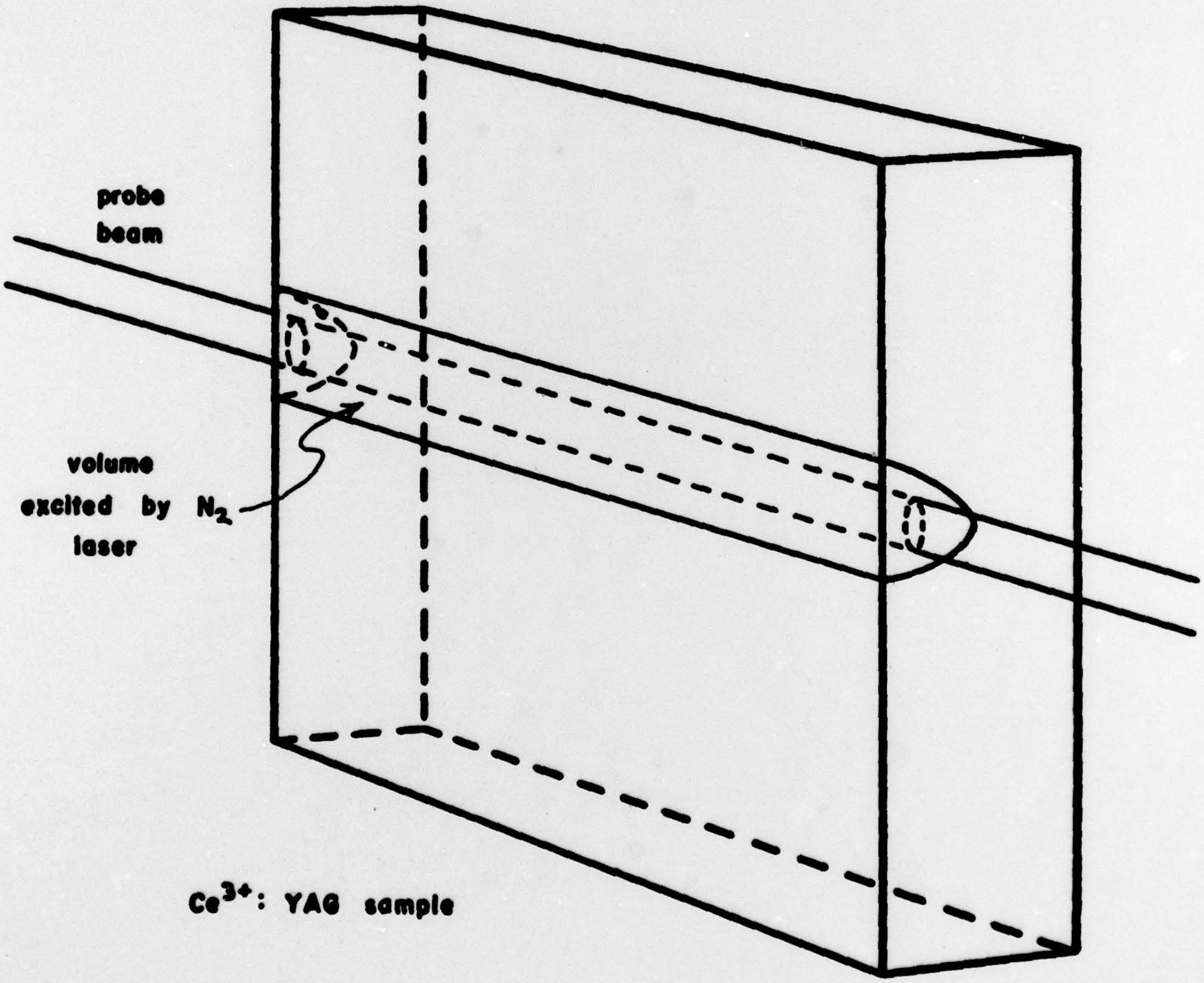
Figure 3. Energy levels for YAG:Ce^{3+}. Insert shows cross section for excited state absorption as a function of photon energy measured relative to the lowest 5d level. Lined region around 19,000 cm^{-1} corresponds to the flourescence photon energy.

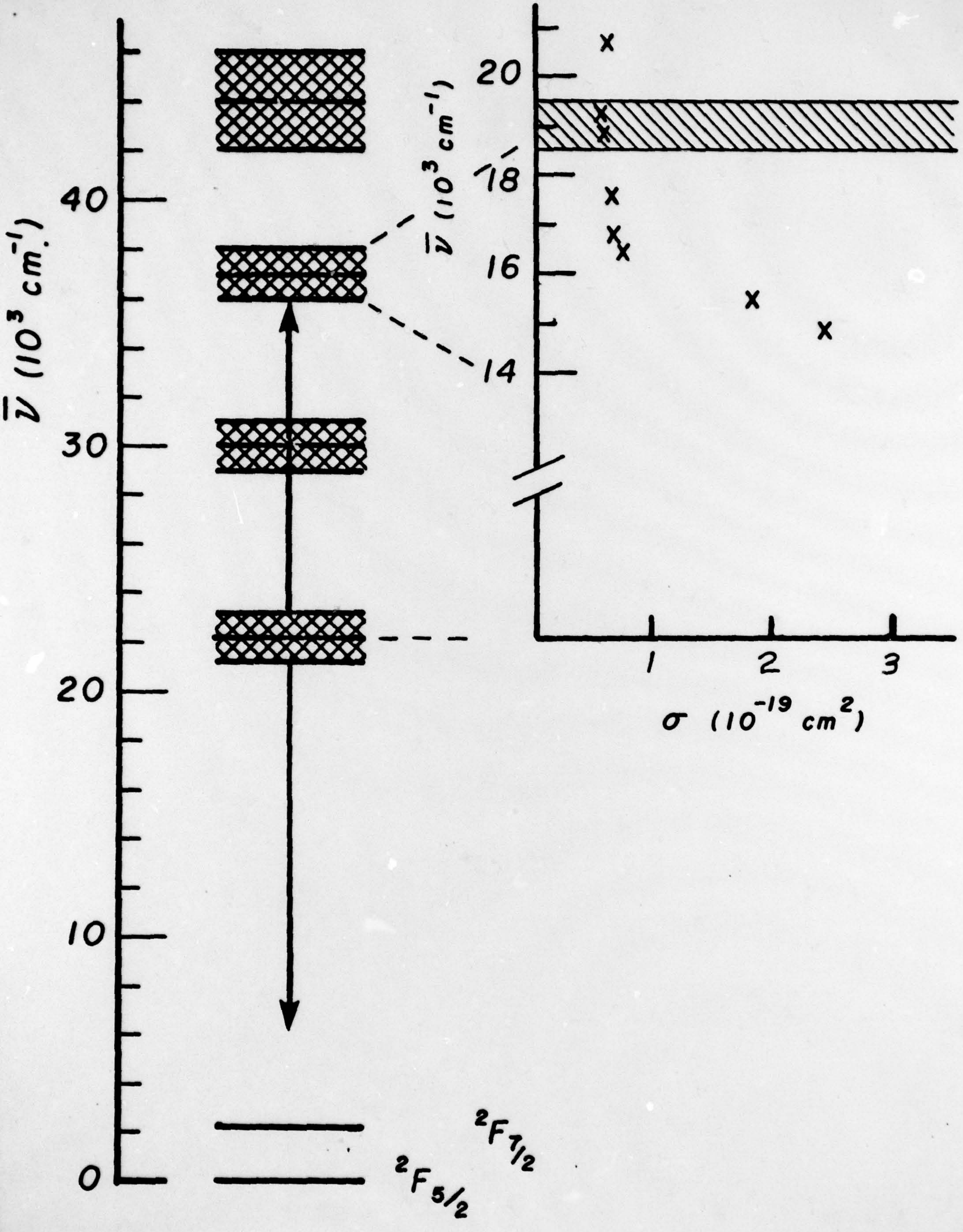
Figyre 4. Magnitude of the short component of ESA at 633 nm as a function of excitation power. Data are the crosses; the solid line is the theoretical fit. Note saturation of absorption resulting from excitation of all Ce^{3+} ions probed. Fit gives ESA cross section of 1.4 x 10^{-19} cm^{2}.

BLOCK DIAGRAM OF APPARATUS FOR EXCITED STATE ABSORPTION MEASUREMENTS

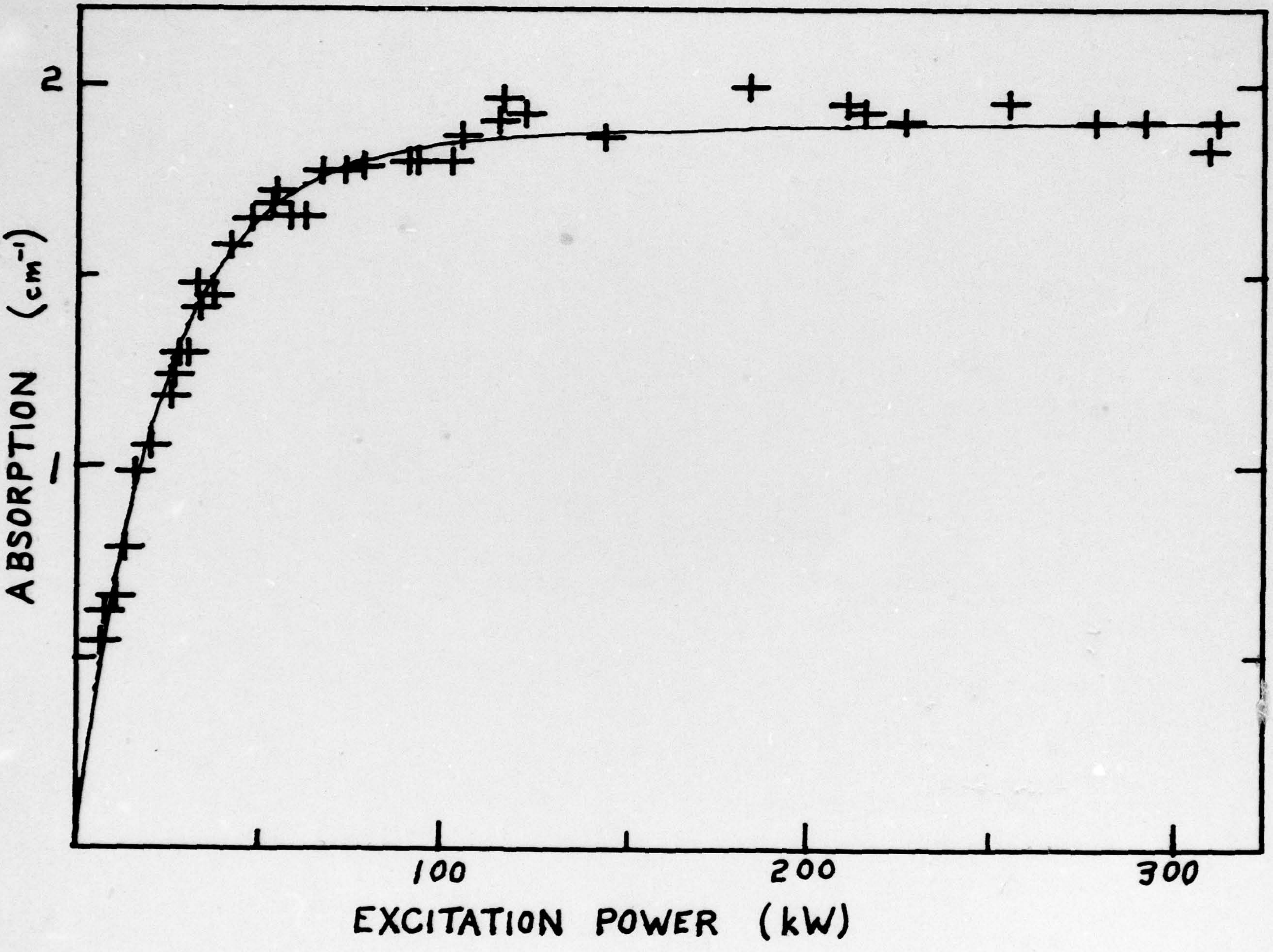


GEOMETRY FOR EXCITED STATE ABSORPTION MEASUREMENTS





$\bar{\nu}$ (10^3 cm^{-1})
40
30
20
10
0
$^2F_{7/2}$
$^2F_{5/2}$
20
18
16
14
1
2
3
σ (10^{-19} cm^2)



2
1
ABSORPTION (cm⁻¹)
100
200
300
EXCITATION POWER (kW)