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EXCITED ATOMS BY SPATIALLY PERIODIC POTENTIALS

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July 1970

LAWRENCE RADIATION LABORATORY
UNIVERSITY of CALIFORNIA BERKELEY

DISCLAIMER

RESONANCE EFFECT OF FAST MOVING EXCITED ATOMS BY SPATIALLY PERIODIC POTENTIALS*

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ABSTRACT

Resonance effects of the fast-moving excited atoms accelerated by an accelerator propagating through spatially periodic potential are described experimentally and theoretically. Atoms or ions propagating through the spatially periodic potential (or field) at the velocity V experience on oscillating frequency v given by $v = V/(2d)$, where 2d is the period of the spatial potential. Thus the resonance effect ranging from rf to microwave frequency can be observed. In addition to the phenomena already observed, some new proposal, concerning the magnetic resonance by spatially periodic magnetic potential are given.

Investigation of excited states of atoms by radio-frequency resonance technique began with the pioneering works by Lamb and Rutherford[1] and Brossel and Bitter.[2] By means of radio-frequency resonance technique, the detailed investigation of excited atoms, not possible by classical spectroscopy, become feasible. Thus, a great advance in atomic physics was made by rf resonance technique.

Because of the high resolution of the radio-frequency spectroscopy, we felt that an application of this technique to the foil excited atoms propagating at high velocity would be of some importance. However, because of the high velocity and short lifetimes of the most of the excited states of atoms and ions, in general, a very high-powered rf generator is required in order to apply the rf resonance technique to beam foil spectroscopy. Because of this difficulty, we searched for an alternative means to achieve the resonance effect without using a rf generator. The foil excited atoms or ions initially accelerated by an accelerator move at relatively high velocity of typically a few percent of the speed of light. The velocity is reasonably uniform and quite directional. Therefore, the atoms propagating with velocity V through the spatially periodic potential whose period is 2d experience on oscillating field whose frequency v is given by $v = V/(2d)$. The required rf power is given by the amplitude of the static spatially periodic field, which can be made extremely high. The frequency one can create artificially for the velocity of atoms of a few percent of the

speed of light ranges from a few MHz to 10 GHz. However, if one passes the beam along the channel direction of the crystalline foil, v seen by the atom could be in a range as high as the vacuum ultraviolet region. Therefore, at least in principle, the usable frequency is from a few MHz to vacuum ultraviolet. At present, no device that can be continuously tuned over and a wide range is available.

We have decided to test the possibility of observing the resonance effect on the excited states of hydrogen atom between the pair of states $n^2S_{1/2}$ and $n^2P_{1/2}$, the Lamb shift, by means of a spatially periodic electric field.[3] We shall show how the resonance effect may be observed by application of a simple time-dependent perturbation theory. A more rigorous description of rf resonance phenomena, using density matrix treatment, is given by Lamb and Sanders.[4] Let the Hamiltonian be $H = H_0 + H_D + H_{rf}$, where H_0 is the Hamiltonian describing the atom, H_D is the damping operator that describes the finite lifetime of the excited states. Therefore, H_D must be diagonal operator given by $H_D|ns\rangle = -\frac{i\gamma_{ns}}{2}|ns\rangle$ and $H_D|np\rangle = -\frac{i\gamma_{np}}{2}|np\rangle$, where γ_{ns} and γ_{np} are the reciprocal lifetimes of the ns and np states respectively. We let H_{rf}, the radio-frequency interaction, be the perturbation term. We wish to solve the Schrödinger equation,

$$i\hbar \frac{\partial \psi(r,t)}{\partial t} = (H_0 + H_D + H_{rf})\psi(r,t). \quad (1)$$

Let us represent the solution by

$$\psi(\underset{\sim}{r},t) = |\underset{\sim}{r},t\rangle$$

$$= \sum_n C_n(t)|n\rangle \exp[-\frac{1}{2}\gamma_{nt} - \frac{i}{\hbar}E_n^{(0)}t]. \quad (2)$$

Since $n^2P_{1/2}$ and $n^2S_{1/2}$ are the most closely lying pair, we approximate eq. (2) by

$$|r,t\rangle = C_{n^2S_{1/2}}(t)|n^2S_{1/2}\rangle \exp[-\frac{1}{2}\gamma_{ns} - \frac{i}{\hbar}E^{(0)}_{n^2S_{1/2}}]t$$

$$+ C_{n^2P_{1/2}}(t)|n^2P_{1/2}\rangle \exp[-\frac{1}{2}\gamma_{np} - \frac{i}{\hbar}E^{(0)}_{n^2P_{1/2}}]t. \quad (3)$$

Now, $H_{rf} = \underset{\sim}{E}(t) \cdot \underset{\sim}{P}$, and we shall approximate this by

$$H_{rf} = \underset{\sim}{E}_0 \cdot \underset{\sim}{P} \cos(2\pi \frac{V}{2d} \cdot)t, \quad (4)$$

where E_0 is the amplitude of the spatially periodic electric field and $\underset{\sim}{P}$ is the electric dipole operator. Let the perturbation be turned on at $t = 0$ to $t = t$, at which the atom propagating with the velocity V enters the region where spatially periodic field is applied and the atom leaves the interaction region at time t later. Let us assume that the atom enters the interaction region in the state $n^2S_{1/2}$. This is a reasonable assumption, since the damping constant of ns is in general, smaller than of any other state, so that the experiment can be arranged in such a way that all the rest of the states decay spontaneously.

Under such an assumption we have

$$C_{n^2S_{1/2}}(0) = 1 \text{ and } C_{n^2P_{1/2}}(0) = 0. \quad (5)$$

Then, to first order, we can write

$$C^{(1)}_{n^2P_{1/2}}(t) = \frac{1}{i\hbar}\int_0^t \langle n^2S_{1/2}|\underset{\sim}{P}\cdot\underset{\sim}{E}_0|n^2P_{1/2}\rangle$$

$$\times \cos 2\pi \frac{V}{2d}t_0 \exp[iW_{sp} - \frac{1}{2}(\gamma_{np}+\gamma_{ns})]t_0 \, dt_0, \quad (6)$$

where $W_{sp} = [E_{n^2S_{1/2}} - E_{n^2P_{1/2}}]/\hbar$, from the standard perturbation theory of the method of variation of constants. Maintaining only the term that exhibits the resonance effect, we have

$$C^{(1)}_{n^2P_{1/2}}(t) \approx (1/i\hbar)$$

$$\times [\langle n^2S_{1/2}|\underset{\sim}{E}_0\cdot\underset{\sim}{P}|n^2P_{1/2}\rangle/\{i(2\pi\frac{V}{2d} - W_{sp})\}]$$

$$\times [\exp\{iW_{sp} - \frac{1}{2}(\gamma_{np} + \gamma_{ns})\}t - 1]. \quad (7)$$

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The probability of finding the atom in the $n^2P_{1/2}$ state is given by

$$|C^{(1)}_{n^2P_{1/2}}(t)|^2 = (1/\hbar^2)$$
$$\times |\langle n^2S_{1/2}|\underset{\sim}{E}_0\cdot\underset{\sim}{P}|n^2P_{1/2}\rangle|^2/-\{4\pi^2(\frac{V}{2d}-\nu_{sp})^2$$
$$+\frac{1}{4}(\gamma_{np}+\gamma_{ns})^2\}\times[\exp\{-(\gamma_{np}+\gamma_{ns})\}t-1].$$
(8)

The time rate of transition is given by

$$\lim_{t\to 0}\frac{1}{t}|C^{(1)}_{n^2P_{1/2}}(t)|^2$$
$$\approx\frac{1}{\hbar^2}\frac{|\langle n^2S_{1/2}|\underset{\sim}{E}_0\cdot\underset{\sim}{P}|n^2P_{1/2}\rangle|^2}{\frac{1}{4}(\gamma_{ns}+\gamma_{np})^2+4\pi^2(\frac{V}{2d}-\nu_{sp})^2},$$
(9)

which agrees with the expression shown by Lamb and Sanders. [4] The rate of emission of quanta from the excited states is given by

$$I(t) = f_{ns}\gamma_{ns}|C^{(1)}_{n^2S_{1/2}}(t)|^2 + f_{np}\gamma_{np}|C^{(1)}_{n^2P_{1/2}}(t)|^2,$$

where f_{ns} and f_{np} are the branching ratio of $|n^2S_{1/2}\rangle$ and $|n^2P_{1/2}\rangle$ states, respectively.

However, we observe the quanta emitted over the finite length, $\Delta\ell$, of the propagating atomic beam. Let the flight time of the atoms in the beam over this length $\Delta\ell$ be T, given by $T = \Delta\ell/V$. Then, the number of quanta emitted in time T is given by

$$I(T) = \int_{t_1}^{t_2} I(t_0)\,dt_0.$$
(10)

Thus

$$I(T)\approx f_{ns}|C^{(1)}_{n^2S_{1/2}}|^2 + f_{np}|C^{(1)}_{n^2P_{1/2}}|^2.$$
(11)

Since $|C^{(1)}_{n^2S_{1/2}}|^2 + |C^{(1)}_{n^2P_{1/2}}|^2 \approx 1$, from the conservation of probability, eq. (11) can be written as

$$I(T)\approx f_{ns} + |C^{(1)}_{n^2P_{1/2}}|^2(f_{np}-f_{ns}).$$
(12)

Remembering that

$$C^{(1)}_{n^2P_{1/2}}\approx\frac{1}{\hbar^2}\frac{|\langle n^2S_{1/2}|\underset{\sim}{E}_0\cdot\underset{\sim}{P}|n^2P_{1/2}\rangle|^2}{\frac{1}{4}(\gamma_{ns}+\gamma_{np})^2+4\pi^2(\frac{V}{2d}-\nu_{sp})^2}$$

$C^{(1)}_{n^2P_{1/2}}\approx 0$ when the $\frac{V}{2d}=\nu\neq\nu_{sp}$ at off-resonance condition, whereas at resonance, when $\frac{V}{2d}=\nu_{sp}$, $C^{(1)}_{n^2P_{1/2}}$ takes the maximum value. The resonance signal, S_{ig}, is given by taking the difference between I(T) on resonance and off resonance. Therefore,

$$S_{ig} = |C^{(1)}_{n^2P_{1/2}}|^2(f_{np}-f_{ns}).$$
(13)

Whether the signal increases at resonance or decrease at resonance is dependent on $f_{np}-f_{ns}$, the difference between branching ratio. If H_α correspond to the $n=3\to n=2$ transition, $f_{3p}-f_{3s}<0$. Therefore, the H_α light intensity decreases at resonance.

Figure 1 shows the experimental arrangement used for the rf resonance detection by a spatially periodic electric field, and Fig. 2 shows the experimental result for the H_α line. The experimental result is in complete agreement with the above theoretical prediction for H_α. For the L_α line, $f_{2s}=0$, since the $|2s\rangle$ state is a metastable state. Therefore $f_{2p}-f_{2s}>0$, so that the L_α line should increase at resonance. This phenomenon is shown in Fig. 3.

In addition to the electric dipole resonance effect, the beam foil spectroscopy technique can be used for radio-frequency magnetic resonance, Hanle effect (zero-field level crossing), and level crossing. In order to observe such phenomena, it is necessary that the excited atoms be either oriented or aligned. Unfortunately, at present, almost no experimental information concerning the alignment of excited states by foil is available. Clearly, more work should be done along this direction.

There is some information available about low-energy protons and hydrogen atoms colliding with stationary hydrogen and helium atoms[5, 6]. In these experiments, for the energy of protons or hydrogen atoms in the range of 10 to 30 keV, the excited atoms are

preferentially more populated in $m_\ell = 0$ than in $m_\ell = \pm 1$, with the beam axis referred to as the quantization axis. Figure 4 shows a typical experimental result of Kraus and Soltysik on the formation and alignment of He in the 3^3D state detected by monitoring the $\lambda = 5876$ Å line corresponding to the 3^3D to 2^2P transition. The percentage polarization π is defined as $\pi = (I_{\parallel} - I_{\perp})/(I_{\parallel} + I_{\perp})$, where $I_{\parallel}$ and $I_{\perp}$ are the intensities of the light polarized parallel and perpendicular to the ion beam. The Hanle effect as well as the level crossing effect can be detected by applying the magnetic field at some angle other than parallel to the beam axis. For example, the application of magnetic field perpendicular to the beam axis creates a coherence between the certain pair of states whose magnetic quantum number differs by $\Delta m = \pm 2$. At the level crossing point where these pairs of states become degenerate, an optical spatially dependent interference effect occurs, resulting in the change of intensity of light or polarization of light emitted.[7] Since this type of phenomena, as well as rf resonance phenomena, is not dependent on the Doppler effect of the emitted light nor on the cascading effect, not only the precise energy level determination, but also the life-time of the excited states can be measured. That Kraus and Soltysik experimentally observed $\pi \neq 0$ for the 3^3D state insures that the level crossing effect can be observed. Figure 5 shows the level crossing signals as well as relevant energy level diagram and experimental arrangement upon proton impact on He, exciting it to 3^3D, used by R. D. Kaul[8] to measure fine structure. These experiments showed that it is very likely that almost any atom can be aligned by ion impact.

Such technique is quite important for the high-field level crossing, since the radius of curvature of the ions propagating through the magnetic field perpendicular to the beam is much less than that of the electron beam of comparable velocity. In the past, the electron beam at the energy slightly above threshold energy of excitation was used to coherently excite the nonresonant (optical) states to observe the level crossing phenomena.

Also, recently, Kaminsky[9] showed that the tensor polarization of deuterons of about 32% can be obtained by a charge-capture process of oriented electrons by passing the deuteron through the channeling direction of magnetized monocrystalline nickel foil. This result indicates that the excited states of deuterium atoms must be highly polarized and this technique should be applicable to other ions passing through the magnetized crystalline foil. The oriented excited atoms produced by such a process can be resonated by a spatially periodic magnetic field. The magnetic resonance phenomenon yields information about g values (g_J and g_F), hyperfine structure, and lifetimes (here the cascading can be completely ignored), since the resonance phenomenon is the property of only a particular pair of states undergoing the magnetic resonance. Even by using the polycrystalline foil, the alignment of the excited state of atoms from the charge-capture process should be possible. It was shown by Hughes[10] that excited states of hydrogen with $m_\ell = 0$ were formed by the $H^+ + e \rightarrow H^*$ process from a gas target. It is not at all surprising that similar phenomena may be observed for other ions by the charge-capture process, either by a foil or a gas target. Again, magnetic resonance induced by spatially periodic potential is possible for states other than those with $L = 0$ and $J = F = 0$ and $1/2$ since only the alignment, not the orientation, can be effected by such a process. On the other hand, when the excited states of an atom are oriented by some technique such as that developed by Kaminsky, orbital

angular momentum L = 0 allows observation of resonance phenomena provided the spin angular momentum S $\neq$ 0.

I believe that these discussion I presented shows that the rf resonance technique may yield a great deal of informations about the excited states of atoms that cannot be obtained by classical techniques.

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*Work done under auspices of the U. S. Atomic Energy Commission, National Aeronautics and Space Administration, and Office of Naval Research.

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FIGURE CAPTIONS

Fig. 1. Experimental arrangements and on electric dipole resonance experiments.

Fig. 2. (a) Relevant energy level diagrams and (b) experimental results of αf signal when the energy of (HHH$^+$) was 400 keV with 2d (a period) = 0.97 cm, corresponding to an effective frequency seen by the moving hydrogen atom at 520 MHz with $E \approx 10 \text{V/cm}$.

Fig. 3. Experimental result of observation of $2\,^2S_{1/2} - 2\,^2P_{p/2}$ energy separation (Lamb shift) by L_α line.

Fig. 4. Alignment and relative excitation cross section of $3\,^3D$ state by proton impact on He atom. (Kraus and Soltysik, Ref. 6).

Fig. 5. Relevant energy level diagram, experimental arrangement and level crossing signal (upper trace) corresponding to $(1, 0) \times (2, 2)$ crossing with NMR calibration mark (lower trace). (R. D. Kaul, Ref. 8).

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Differentially pumped bombardment chamber containing helium

Beam collector

Chamber window

Crossing magnetic field up out of paper in this region

Beam drift tube

40 keV H₂⁺ beam

$40 \text{ keV } H_2^+ \text{ beam}$

Lens

Polarizer

Monochromator

5875A

Photomultiplier
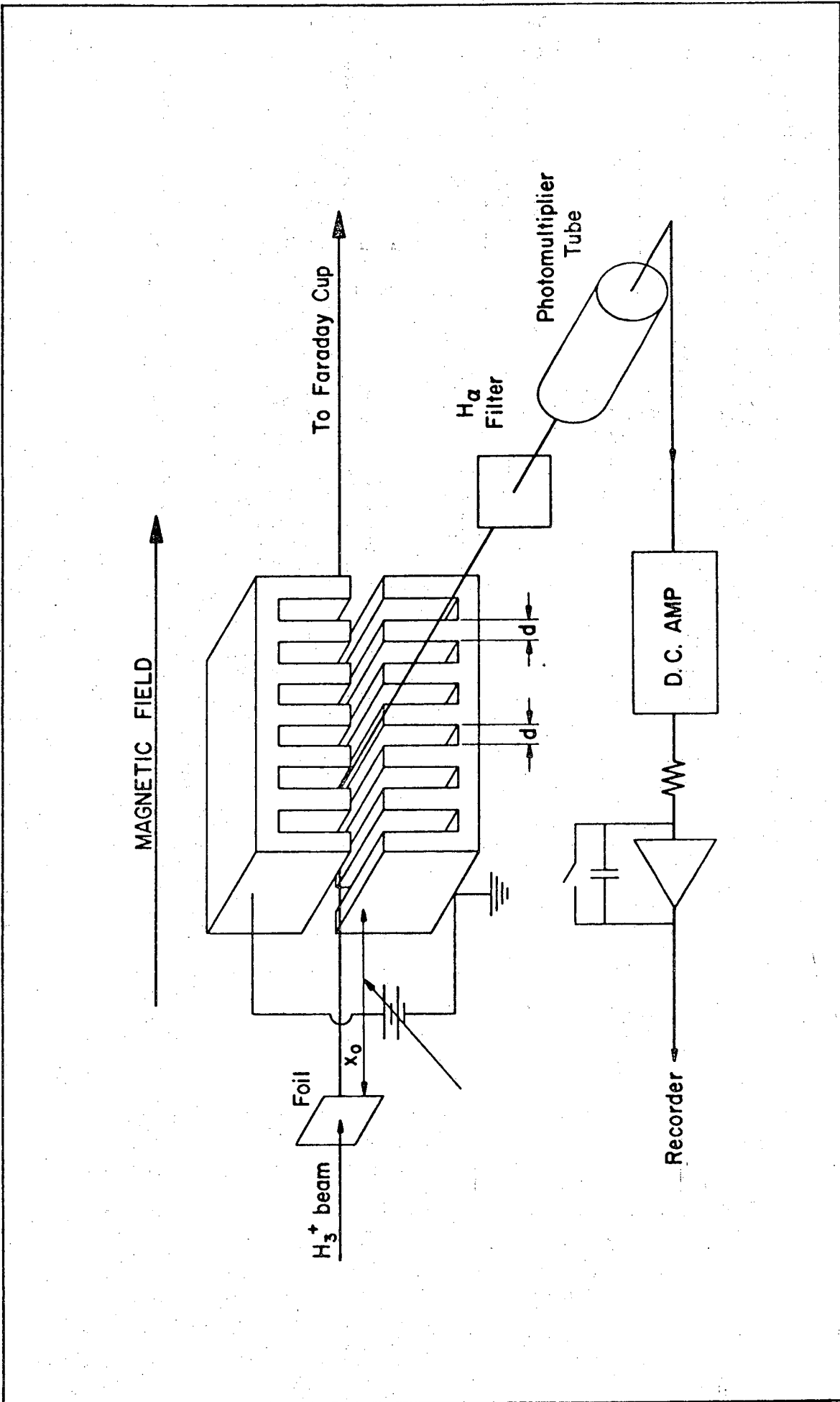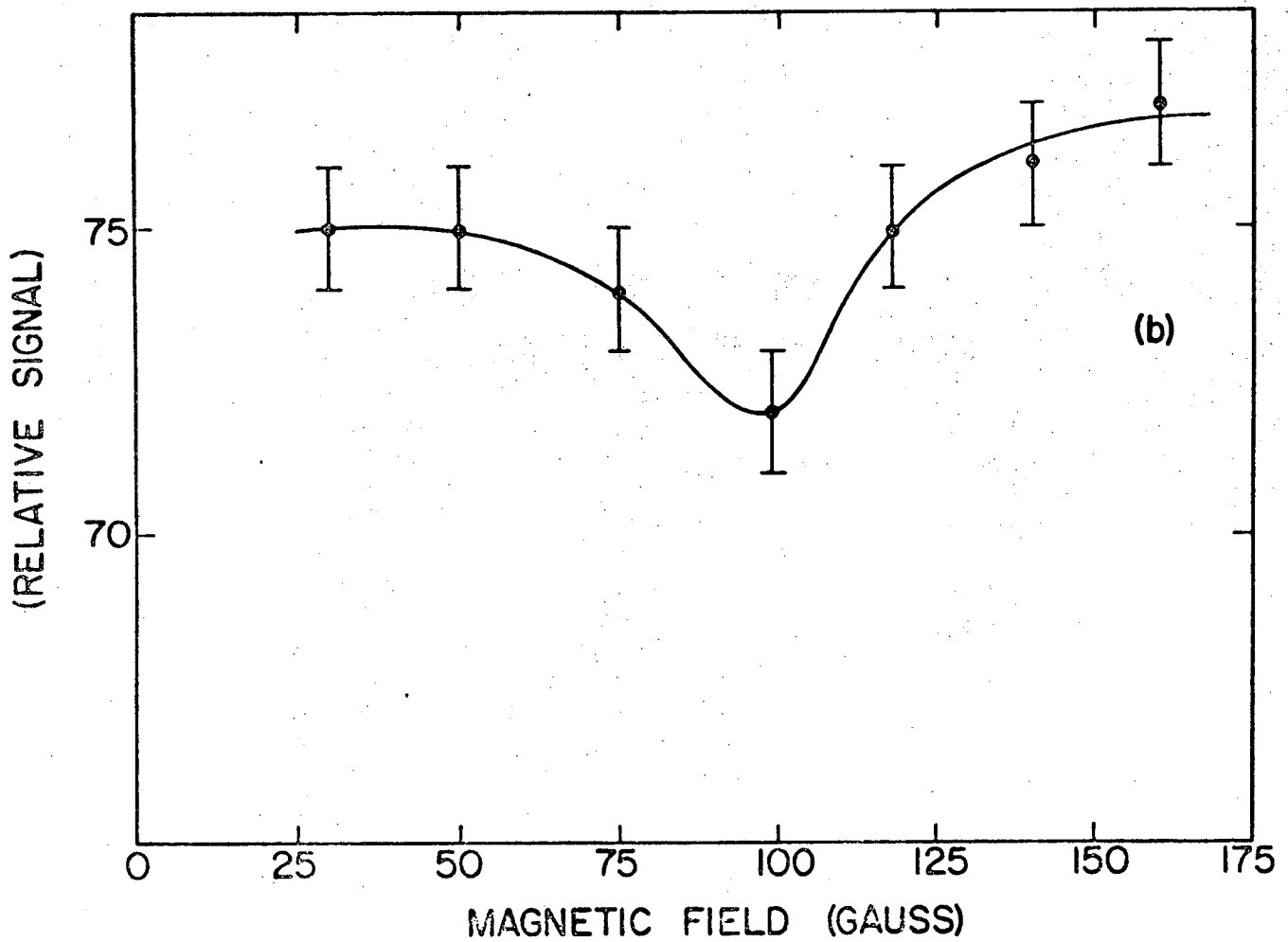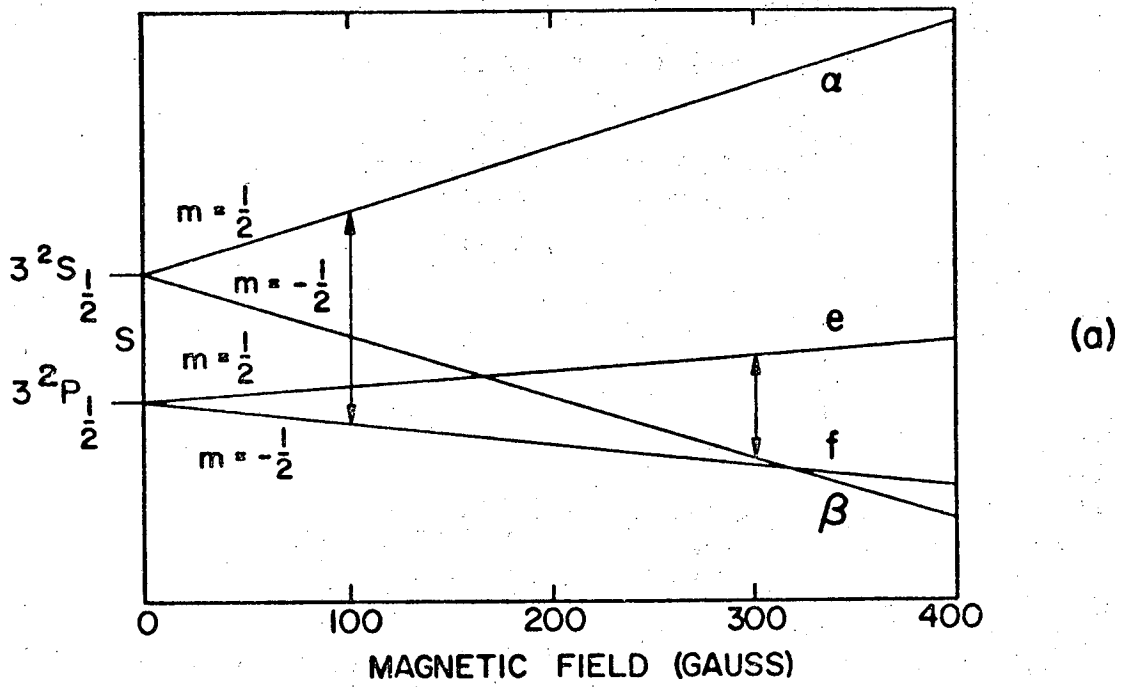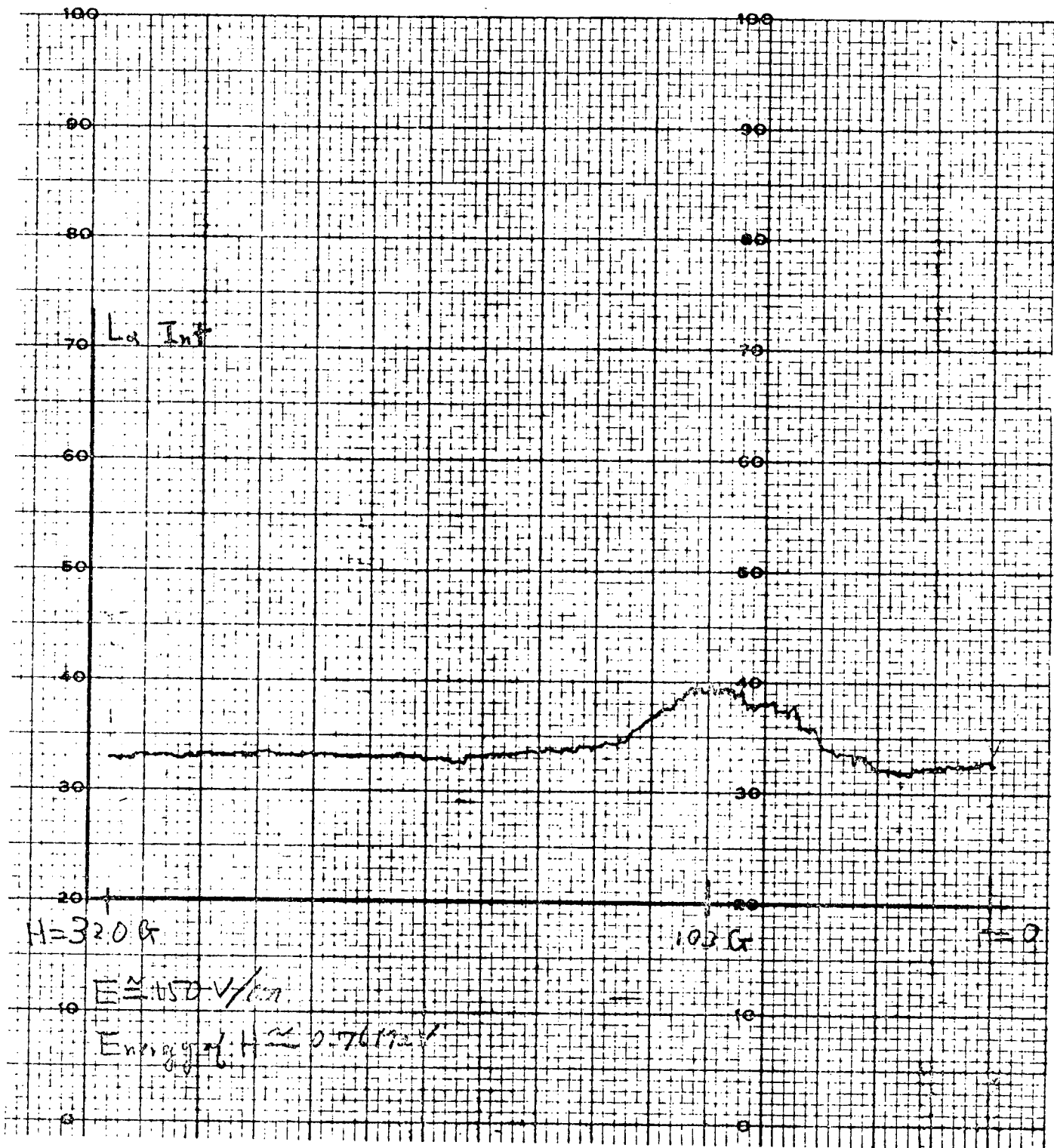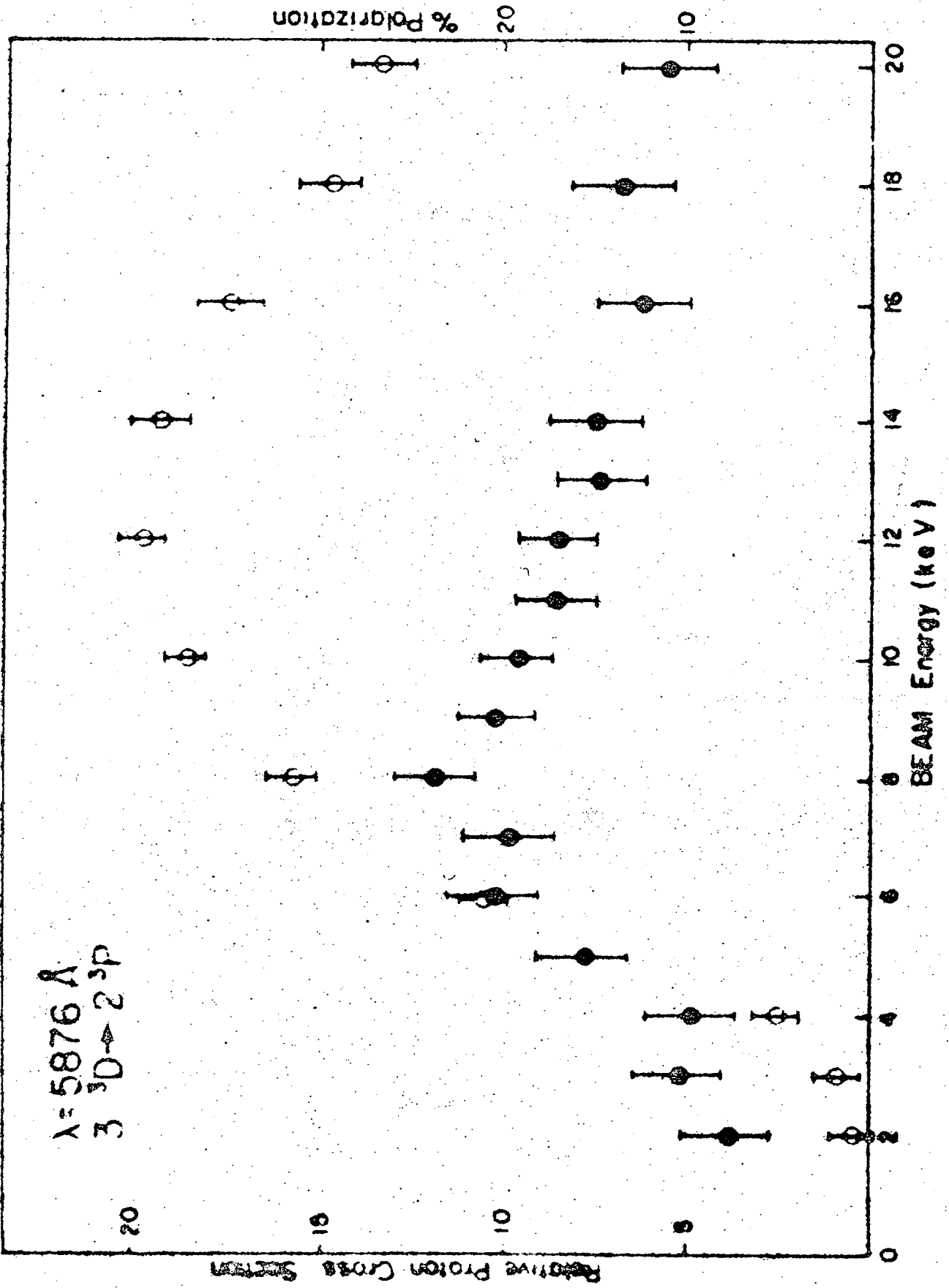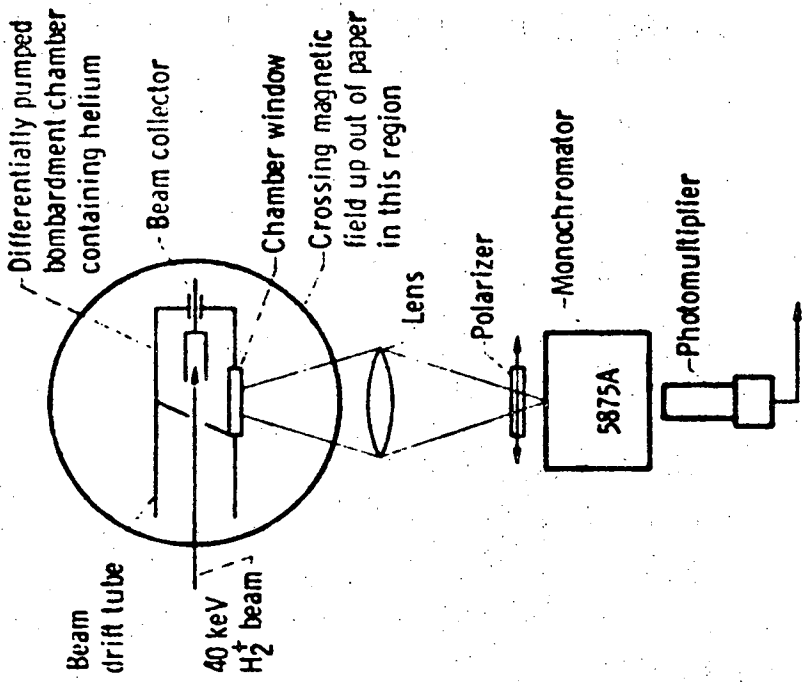
FIG. 2. Apparatus schematic.

Fig. 5

Energy, MHz

2000

1500

1000

500

0

-500

0   100   200   300   400   500   600

Magnetic field, gauss

J = 1

J = 2

J = 3

(1, 1)x(3, 3)

(1, 0)x(3, 2)

(1, 0)x(2, 2)

(1, -1)x(2, 1)
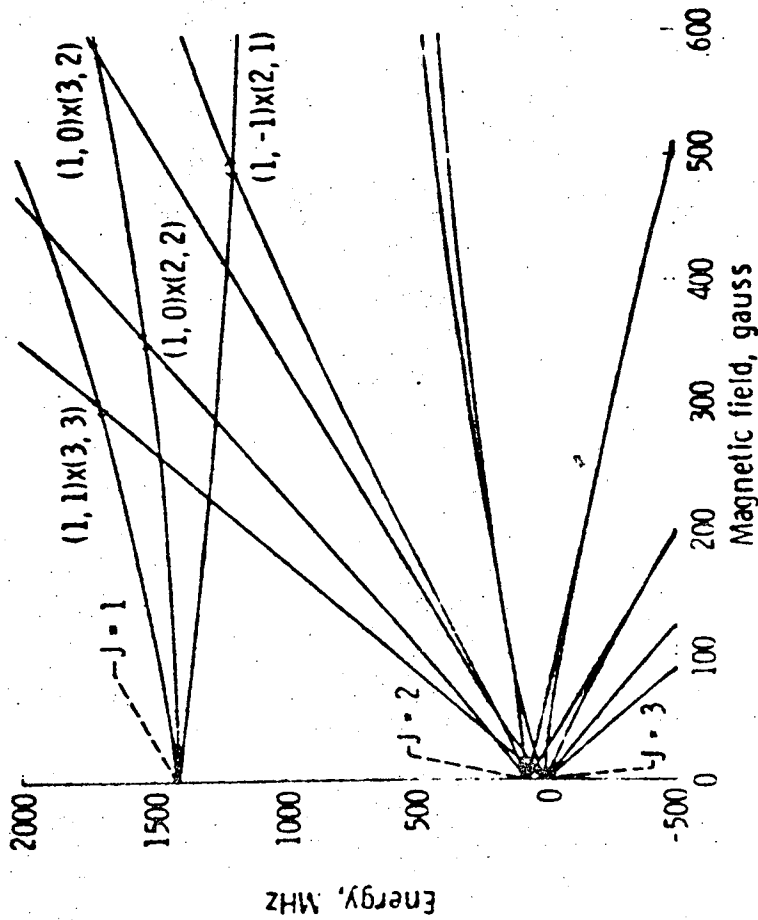
OPTICALLY EXCITED HANLE EFFECT

50 FULL SCALE

$\lambda 2537 \text{ Å } {}^1S_0 - {}^3P_1 \text{ HgI}$

LIGHT SOURCE

LINEAR POLARIZER

TO DETECTOR

H(GAUSS) ⟶

POLARIZED LIGHT FROM EXCITATION OF Hg BY Ar⁺

$Ar^+ \ E \approx 1/2 \text{ MeV}$

$I = 5 \mu A$

2000 COUNTS/FULL SCALE

DARK CURRENT ≈ 10%

$\lambda 2537 \text{ Å } {}^1S_0 - {}^3P_1$

TO MONOCHRONOMATOR

H(GAUSS) ⟶

Fig. 6