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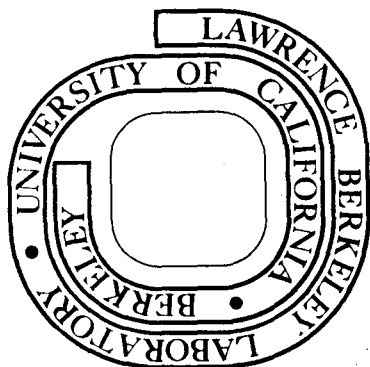
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RADIO-FREQUENCY ELECTRODELESS DISCHARGE LAMPS
FOR USE IN MAGNETIC FIELDS*

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ABSTRACT

There is a demand for radio-frequency-excited electrodeless discharge lamps, for research or instrumental purposes, which operate well in strong magnetic fields. The characteristics and operation of such lamps are described. Magnetic scanning measurements using the resonance radiation of Hg, Cd, and Pb lamps are discussed in some detail, but the lamp design and excitation configuration employed appear to be applicable to a much wider range of elements. An excitation arrangement which reduces the deterioration of the lamp envelope by the discharge is discussed. Further lamp life enhancement is achieved by a rejuvenation procedure. The lamps are particularly suited for use in a background-correcting atomic absorption spectrometer, or as a polarized light source tunable near a resonance line.

I. INTRODUCTION

Electrodeless discharge lamps offer advantageous properties for atomic spectroscopy research, and are often so employed. These properties include intense resonance lines, absence of self-reversal, and stability.¹ Particular virtues of the electrodeless discharge are its ability to operate with an extremely small quantity of metal or isotope and its applicability to many elements. Consequently, the electrodeless discharge is particularly suited to the Zeeman scanning technique.² Zeeman scanning is accomplished by shifting Zeeman components of an isotope lamp with an external uniform magnetic field, and selecting particular components with characteristic polarization. This effectively produces a tunable polarized light source, with a useful tuning range near an atomic resonance line. Electrodeless discharge lamps for such research use are generally operated at microwave frequencies, which are claimed to favor long lamp life and discharge stability.³ However, for research⁴ or instrumental⁵⁻⁸ purposes it is often desirable to use radio-frequency excitation. Lately we have developed a trace-element detector, the Isotope-shift Zeeman-effect Atomic Absorption (IZAA) spectrometer,⁶⁻⁸ based on atomic absorption (AA), which does not suffer the inability of many conventional AA spectrometers to distinguish resonance absorption from broad-band absorption. IZAA detectors require lamps which produce intense non-reversed resonance lines which are in some cases emitted by isotopes of elements. These lamps must be long-lived and must be operated in strong magnetic fields. It has been demonstrated with alkali-metal lamps that long life and stability

are not incompatible with radio-frequency excitation.⁵ Since a radio-frequency exciter is smaller, lighter, and less expensive than one operated at microwave frequencies, we chose to investigate this alternative.

Difficulties in operating rf discharges in magnetic fields have been reported.^{4,9} These difficulties include instability, large self-reversal, and orders-of-magnitude loss of intensity in the resonance lines when the magnetic field was applied. A radio-frequency-excited potassium resonance lamp has been operated at kilogauss fields, but its properties depend strongly on the geometry of the lamp, the exciting coil, and the amount of inert gas and metal used to fill the lamp.⁹ A sodium lamp, with a modulated light intensity, has been operated in a magnetic field at very low radio-frequencies.⁴ We have also found that a mercury lamp, constructed and excited in the manner suggested by Shernoff,¹⁰ operates well in high magnetic fields. However, such a small spherical lamp was found not to be satisfactory for other elements. A program was then initiated to investigate the success of rf discharge of several elements in high field strengths as a function of lamp envelope shape and rf excitation mode. Our initial interest has been directed toward the elements mercury, cadmium, and lead. These metals are of primary environmental concern due to their hazardous potential even in trace quantities. The IZAA detectors which use these lamps operate on the principle outlined below.⁶⁻⁸

Consider a discharge of a single (even) isotope of an element whose excited (resonance) level is split into a classical Lorentz triplet by an external uniform magnetic field. By choosing the correct isotope, one Zeeman component of the resonance line, with characteristic polarization,

can be centered on the Lorentz-broadened and -shifted absorption profile of atoms in air, while other Zeeman components, with different polarization, can be shifted to the wings of this absorption profile. With this configuration, one component monitors both resonant and nonresonant (broad-band) absorption produced by molecular interferences, while the other components monitor this nonresonant absorption alone. The monitor and reference beams are distinguished by their polarizations, which are characteristically associated with the Zeeman effect. The difference in absorption, normalized to the incident light level, provides a measure of the true atomic absorption.

Clearly, the requirements set by this detection technique on the characteristics and operation of the light sources are substantially identical to those found in atomic physics research, and noted above. Additionally, it is desirable in an instrument which uses multiple light sources to have lamps similar in size and shape for each element. For a large range of linearity, as well as high sensitivity, the lamps should emit intense resonance lines with oscillator strengths f in the range 10^{-3} to 1. The life of the lamps should be as long as possible, the construction of the lamps should not depend critically on any parameters, and the lamps should be stable and have low noise. Most, if not all, of these requirements are met by the lamps described below.

II. LAMP CONSTRUCTION

Our investigations show that for good operation in high magnetic fields, the shape of the envelope, the mode of excitation, and temperature control are of far more importance than any novel procedures of lamp construction. The quartz envelope of each lamp is prepared and

filled in a conventional manner.^{1, 11, 12} After the envelope is blown, it is sealed to an oil-diffusion-pumped vacuum system and baked at 1000°C for 4 hr. After cooling, argon is discharged in the envelope and then pumped out, to further clean the walls.¹¹ Distilled elements like Pb and Cd, when used in natural abundance, are driven with a flame into the envelope until an opaque coating covers about one-sixth of the surface area (see Fig. 1). The quantity of metal is not at all critical; the amount can be reduced more than an order of magnitude, but relatively large amounts are currently used in the hope of enhancing lamp life, since some of the metal is eventually embedded in the quartz envelope. Isotope lamps are filled with a small measured quantity of the isotope, using a procedure similar to that outlined by Shernoff.¹⁰ Both Cd and Hg are discharged in elemental form. The vapor pressure of Pb is relatively low at moderate temperatures, so iodine is also driven into the Pb lamp and reacted with the metal until a color change is observed. The remaining free iodine is then driven from the lamp. Each lamp is filled with 2 Torr of research-grade argon, used as a carrier gas. Although the lamps function best when some of the parameters are optimized, and care is required during the filling process, we have not found either detailed envelope preparation¹³ or precise control of the filling material⁹ to be necessary. After standardization of the procedure every lamp constructed has been successfully discharged.

After much experimentation we settled on a "dumbbell" shape for the quartz envelope of the lamps. This shape is one of those discussed by Tolansky¹⁴ and has been found satisfactory for the six elements we have investigated to date. Since the lamps were to be situated in a

narrow gap of a permanent magnet (field strengths to 19 kG have been used), and an unreversed line shape was desired, the portion of the lamp emitting the useful light was made small in cross section. It is difficult to maintain a stable discharge in a capillary due to local pressure build-up, so bulbs were placed at both ends to serve as expansion chambers. The free movement of the metal in a lamp with a single bulb at one end was found to be inhibited. The exception, mercury, was discharged successfully in every envelope shape tested. Discharge uniformity follows from the cylindrical symmetry of the lamps (see Fig. 1). The discharge can be made relatively strong in the capillary tube. Dimensions again are not critical, but the lamps in Fig. 1 have a capillary made of 3/32-in. inside diameter tubing about 1 in. long. The discharge continues into the bulbs, where it spreads to fill the whole cross section. The bulbs are 1/2 in. or more long and 1/2 in. in diameter. The tip left by seal-off serves as a reservoir for the metal.

III. LAMP OPERATION

The lamps have been successfully excited at microwave frequencies, but for our purposes a radio-frequency discharge was desirable. Figure 2 shows the excitation and heating arrangements currently used for the lead and cadmium lamps; the mercury discharge is excited in the same manner, but no heating is required. The oscillator is a conventional push-pull type based on transistors or electron tubes; the higher power tube version operated at 300 V and 80 mA per electron tube. The mercury lamp has also been operated with lower power dual transistor circuits. The single-ended excitation circuit illustrated in Ref. 10 was also adequate for Hg, although discharge stability was less satisfactory.

The frequency of excitation is not at all critical, discharges having been struck over the frequency range 50-120 MHz. However, some care in decoupling the lamp heating coils from the excitation coils is necessary. The arrangement shown in Fig. 2 has proven satisfactory. This decoupling scheme is aided by the basic symmetry of the exciting system. With insufficient decoupling, or operation at very low power, the discharge is extinguished at the center of the capillary, i. e., in the magnet gap, if a magnet is used.

It was possible to approximately ascertain lamp envelope temperatures after some period of operation in the case of Hg, which requires no external heating. The capillary was found to be near room temperature, while the bulbs will severely burn the skin, if touched. The heating of the bulbs could occur through rf heating of the quartz or ion bombardment of the envelope: the latter is suspected. This is borne out by observing the darkening of the Hg lamp envelope following long continuous operation. Such darkening occurs only in the capillary, using the excitation method of Fig. 2. We interpret the relative clarity, and high temperature, of the bulbs to indicate clean-up by ion bombardment sputtering,¹⁵ which might be less effective in the capillary due to the distance from the rf coils, and to the presence of the magnetic field. Further, the metal tends to migrate to the coolest portion of the lamp. The discoloration is not dependent on the quality (i. e., ultraviolet transmission) of the quartz, so the formation of color centers is not the probable cause. Also, the darkening is not necessarily uniform, unlike darkening produced by light flux.

It is noteworthy that the mode of lamp excitation has more effect on the envelope discoloration than the parameters of the lamps themselves. With the same oscillator, the coil is replaced by capacitive excitation if small loops at high rf potential are mounted at the ends of the capillary tube. This mode of excitation causes black deposits to rapidly build up near, but not under, the wire loops. This effect has been discussed by Tolansky.¹⁴ It is thought to also be associated with sputtering. During 2000 hr operation the deposits increased, and eventually the discharge shifted from Hg to argon. We interpret this to mean that the deposits are related to Hg embedded in the quartz by the discharge, which is excited by high electric field strengths near the electrodes. These deposits could not be removed by subsequently heating the envelope with a torch, even to the softening temperature. Similar black deposits have been observed in Cd and Pb lamps, particularly if the metal is not kept from the capillary region by a temperature gradient. The interpretations of the various observations discussed above are not necessarily inconsistent. The degree of ion bombardment in the capillary and in the bulbs, with the two different excitation methods, is markedly different, and in the latter case metal ions may play the dominant role.

The lamps pictured in Fig. 1a show envelope discoloration of a different magnitude, if not of a different type. The Hg lamp shows practically no deterioration after 2800 hr of operation. During 550 hr of operation, deposits formed in the capillary of the Pb lamp. Most of these deposits were removed by subsequent heating of the envelope with a flame (Fig. 1c) to make the lamp re-usable. The Cd lamp (Fig. 1a, center) shows a different instance. This particular type of coating

seems to be associated with failure to maintain a sufficient temperature gradient to the reservoir. The near-uniform brown deposit was a thin layer of metal, which was later completely driven off with a torch before lamp operation continued. Figure 1b shows the same lamp after further discharge, with the metal again driven to one end.

It is often observed that the metal deposition occurs in the capillary primarily between the heater wires, despite these wires being near ground potential. When the lamp is operated in a magnet a relatively cool area naturally occurs near the pole tips. One notes that the most persistent discoloration of the Pb lamp occurs in this area (Fig. 1c). Additional insulation about the pole tips, heating of the exposed area, and a temperature gradient to ensure that the reservoir is the coolest portion of the envelope surface should go far to alleviate this residual deterioration.

Under optimum conditions of operation the life of a mercury lamp is demonstrably greater than 3500 hr. No light loss or additional noise in the resonance light was observed after 2000 hr. We have discharged Cd and Pb lamps usefully for 1000 hr without using the rejuvenation procedure. Neither the life of the lamps nor the ease of discharge is appreciably affected by the presence of the magnetic field. This is in strong contrast to the experiences of others, who report substantial intensity loss when a field is applied to otherwise satisfactory lamps of their design.

IV. MEASUREMENTS

Figure 3 is a diagram of an apparatus used to obtain the absorption profiles of atoms in vacuum and in an atmosphere of argon. The Zeeman scan of the σ components of the resonance radiation was accomplished by applying a variable uniform field H_0 to the lamps.² Unlike the mode of operation pictured in Fig. 2, this field was uniform over the whole lamp envelope. A particular component of the radiation is selected on the basis of its polarization. The circularly polarized light of one σ component, emitted parallel to the field direction, is converted to linearly polarized light by the quarter-wave plate. The light then passes through the absorption cell and is selected by the linear polarizer oriented to pass light of this polarization to the 1P28 photomultiplier detector. By changing the magnetic field applied to the lamp, the σ component is scanned linearly across the absorption profile of the atoms in the cell.

For measurements in the variable field, the excitation was transmitted from the oscillator to the coil surrounding the lamp by a pi coupling network. Variation in resonance light intensity as a function of field strength in the range 0-18 kG was typically less than a factor of 2; the normalized transmitted intensities for scans of Hg, Cd, and Pb vapors are plotted in Fig. 4. Relatively large atom densities were used to better show the structure of the absorption.

The scan of the natural Hg absorption in an atmosphere of argon was made with a lamp containing primarily isotope 204; however, the isotopic purity was at best 70%. It is clear that magnetic field strengths in excess of 12 kG are needed to Zeeman-split the σ components of the lamp to the wings of the absorption profile for use in an IZAA apparatus.

The Doppler width at half maximum is $2\Delta (\ln 2)^{1/2}$ for the absorption in vacuum, with Δ about 1 GHz, and the full width at half maximum (FWHM) of the emission line of an uncooled isotope lamp folded into the absorption was about 3 GHz. The Cd 3261-Å absorption was scanned with the natural mixture, since the isotope shift in Cd is relatively small. The Doppler width for the absorption is expected to be about 1.9 times that for Hg; the combined FWHM for the lamp emission and the absorption can be estimated at ~ 4 GHz from Fig. 4b. Evidence of reversal in the Cd 2288-Å resonance line appeared when the central portion of the lamp was insufficiently heated; substantial accumulation of the metal in this region occurred under these conditions. With better relative heating in the central region, the line width of the emission was still self-broadened (~ 7 GHz FWHM), based on line-crossing measurements.¹⁶

The emission line of Pb at 2833 Å also was relatively broad but showed no signs of reversal either in line-crossings or in spectra observed in 17th order on a 3-m Jarrell-Ash spectrometer.¹⁷ The absence of reversal is no doubt due to the use of PbI_2 ; the residual width may be attributed to pressure-broadening by the vapor, or by free iodine remaining in the lamp. The emission width was ~ 6.5 GHz, compared with the Doppler width constant Δ for the absorption: 1.6 GHz. The absorption scans of natural Pb and Cd in argon (Fig. 4b, c) show that fields of about 8 kG are adequate to transmit the Zeeman-split σ components of the lamp.

The detected intensities of the resonance lines of Pb, Cd, and Hg in an IZAA apparatus are comparable, and strong enough to force the

operation of the photomultiplier at substantially reduced gain to avoid saturation. A crude estimate of the resonance-line power for the Hg transition at 2537 Å was made in the following way. The light source was a Hg lamp driven by a single-transistor oscillator operating at 4W supply power. The detector was a shielded 1P28 photomultiplier tube, 1 m from the lamp, with an interference filter placed before the photocathode. The transmission of the interference filter was 12% at 2537 Å. The light passed through a 1-mm-diameter pinhole placed before the lamp. The absolute sensitivity calibration of the 1P28 was assumed. On the basis of the detected photocurrent and typical 1P28 parameters, the power emitted from a 1/8-in. length of the capillary was calculated to be about 1 mW, sufficient for optical pumping purposes.¹⁰ For normal operation, using the circuit of Fig. 2, 40 W of supply power are available.

V. SUMMARY

The characteristics and operation of general-purpose radio-frequency electrodeless discharge lamps, which operate well in strong magnetic fields, have been described. Preparation and filling of the lamps is performed in a conventional manner. The shape of the lamp envelope, the manner of excitation, and temperature control were found to be the main factors for operational success. The resonance radiation has been found to be fairly intense, unreversed, and stable when the lamp temperature is adequately controlled. Observed long-term envelope deterioration has been arrested by a rejuvenation process. Recent progress indicates that further improvements in lamp life and stability should be forthcoming. Current plans include further research on lamp envelope and

filling characteristics, and extension to other elements. The method appears applicable to many elements; at this time six metals have been successfully discharged: Hg, Cd, Pb, As, Se, and Zn. The behavior of the lamps in a magnetic field is similar, and the field may be uniform over the whole lamp envelope or be applied only to the central portion of the lamp.

ACKNOWLEDGMENT

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Footnote and References

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17. We are indebted to R. D. McLaughlin for this measurement.

Figure Captions

Fig. 1a. Pairs of Hg, Cd, and Pb electrodeless discharge lamps (left to right). In each pair the lamp at the left has not been operated, while the lamp at the right has been discharged for a measured time interval. The lamps have been chosen to illustrate different forms of envelope deterioration. The Hg lamp was discharged for 2800 hr, the Cd lamp for 250 hr, and the Pb lamp for 550 hr. The discoloration of the lamp envelopes is exaggerated by the back-lighting used in the photograph. Figure 1b shows the same pair of Cd lamps again. The discharged lamp (bottom) has been rejuvenated by heating with a torch, to drive the metal to one end. It was discharged again for 125 hr (375 hr total) and again heated with the torch. Figure 1c pictures the same pair of Pb lamps, following the torch heating procedure, but without further discharge. The scale in each photograph is calibrated in cm.

Fig. 2. An excitation circuit and heating arrangement used to discharge the lamps. The heater coils are used for Cd and Pb, but not for Hg, lamps. The leads labeled A-A, B-B go to dc heater supplies. The chokes labeled C are wound on Teflon forms. They serve to isolate the heater coils from the rf excitation coils of the oscillator tuned circuit. The inductance of this circuit is the coil around the lamp. The oscillator is conventional, with a typical excitation frequency of 85 MHz.

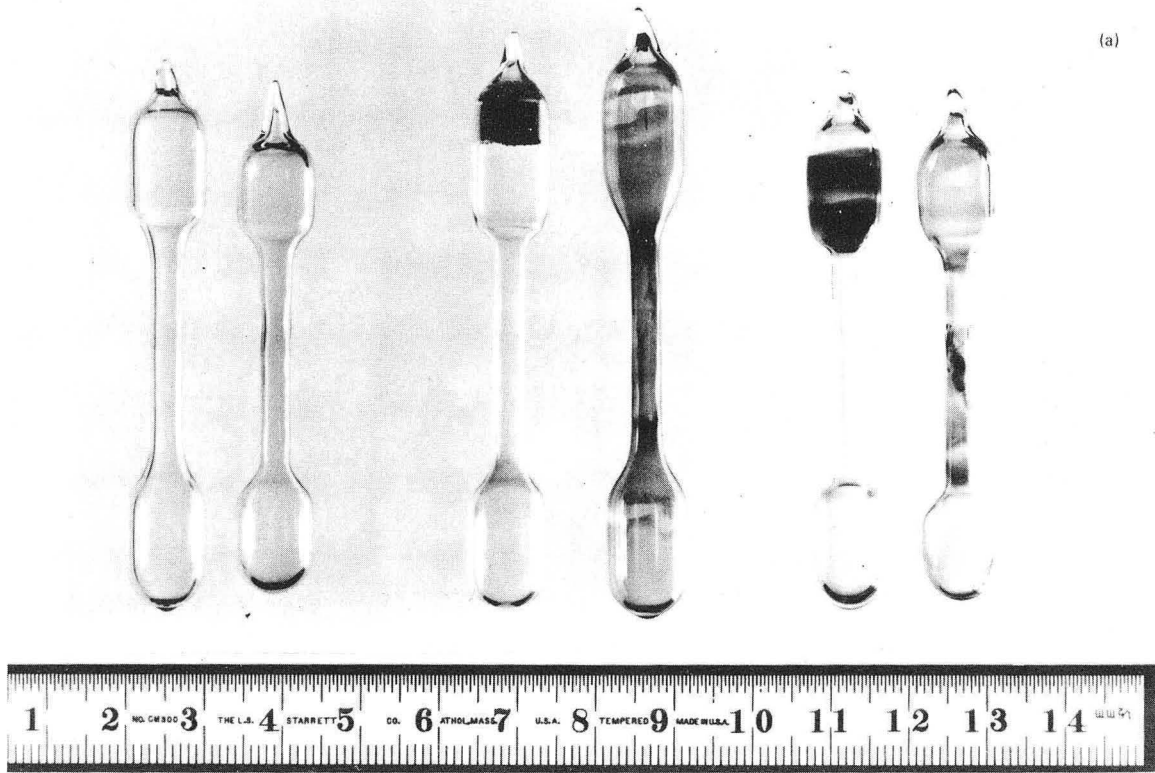
Fig. 3. A diagram of an apparatus used for Zeeman-scanning measurements on atoms in vacuum, or in an atmosphere of argon. The lamp is discharged in a uniform variable magnetic field H_0 . The

quarter-wave plate is a slab of stressed quartz.

Fig. 4a. An absorption profile of natural Hg in an atmosphere of argon, made with a ^{204}Hg isotope lamp scanned with the apparatus of Fig. 3. The transmission is normalized to the light intensity in the absence of the cell, which varied slightly with magnetic field strength. The 2537 \AA transition was used.

Fig. 4b. Zeeman scan of the absorption profile of natural Cd in vacuum, and in approximately one atmosphere of argon, made with a natural Cd lamp using the 3261-\AA intercombination line.

Fig. 4c. Zeeman scan of the absorption profile of natural Pb in argon, using resonance radiation at 2833 \AA from a ^{204}Pb isotope lamp. Again, the argon pressure is only approximate.

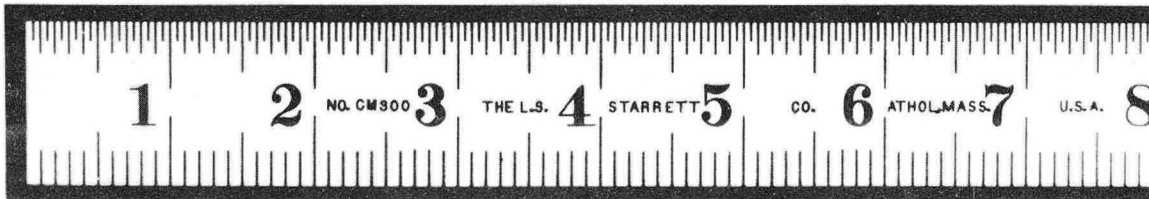
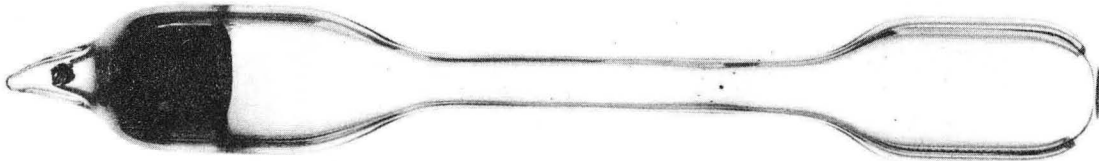


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Fig. 1a.

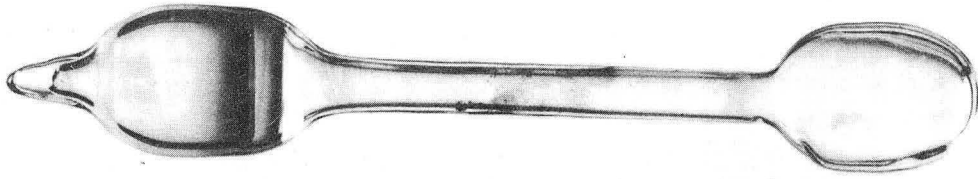


(b)

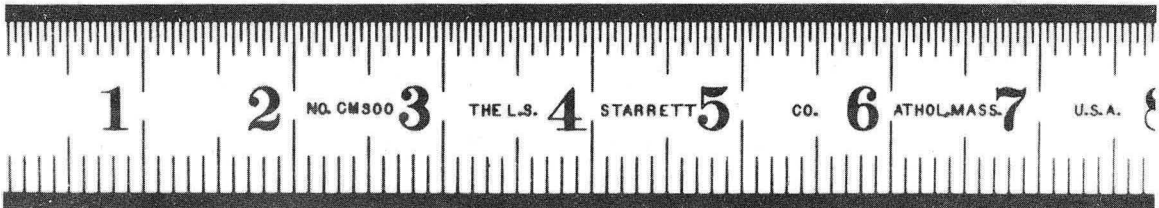
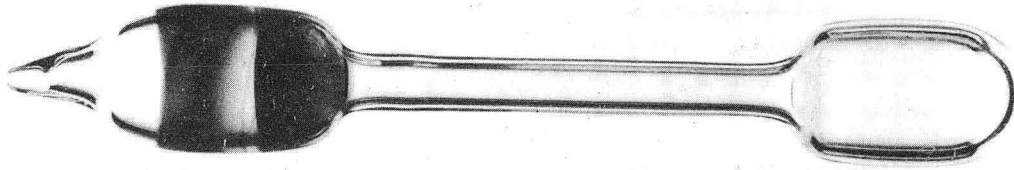


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Fig. 1b.

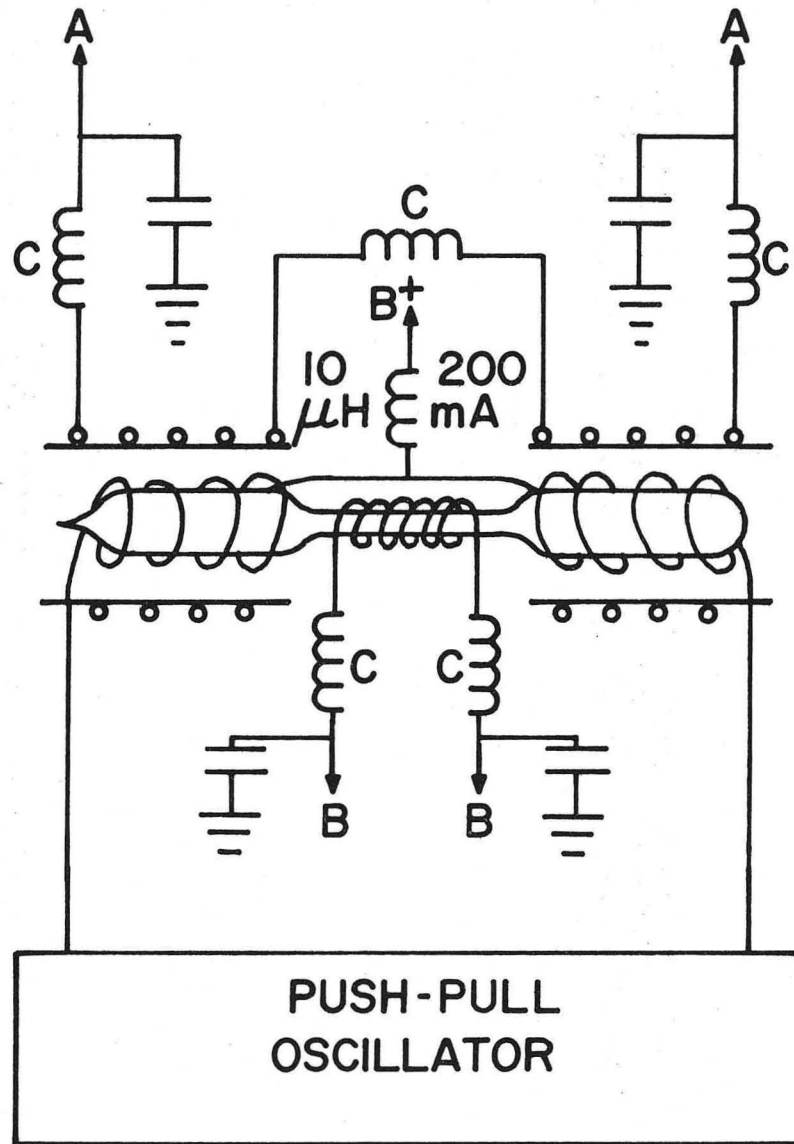


(c)



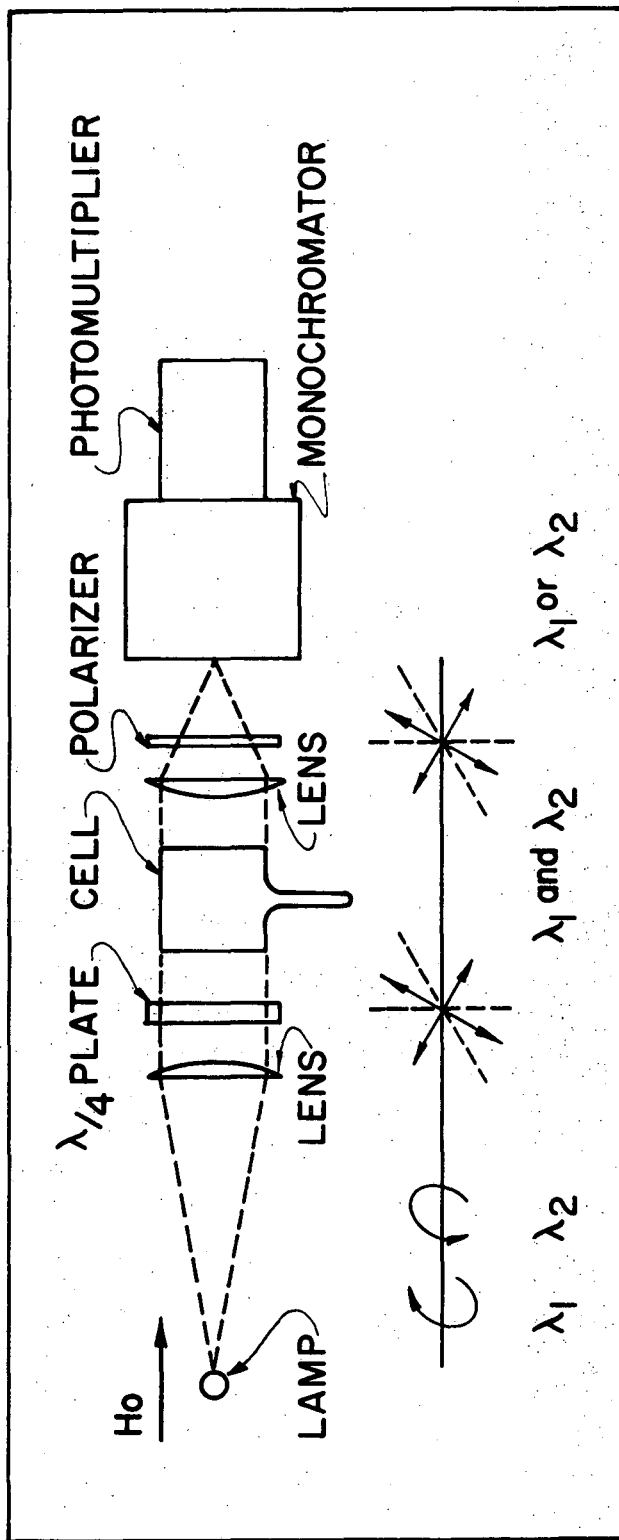
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Fig. 1c.



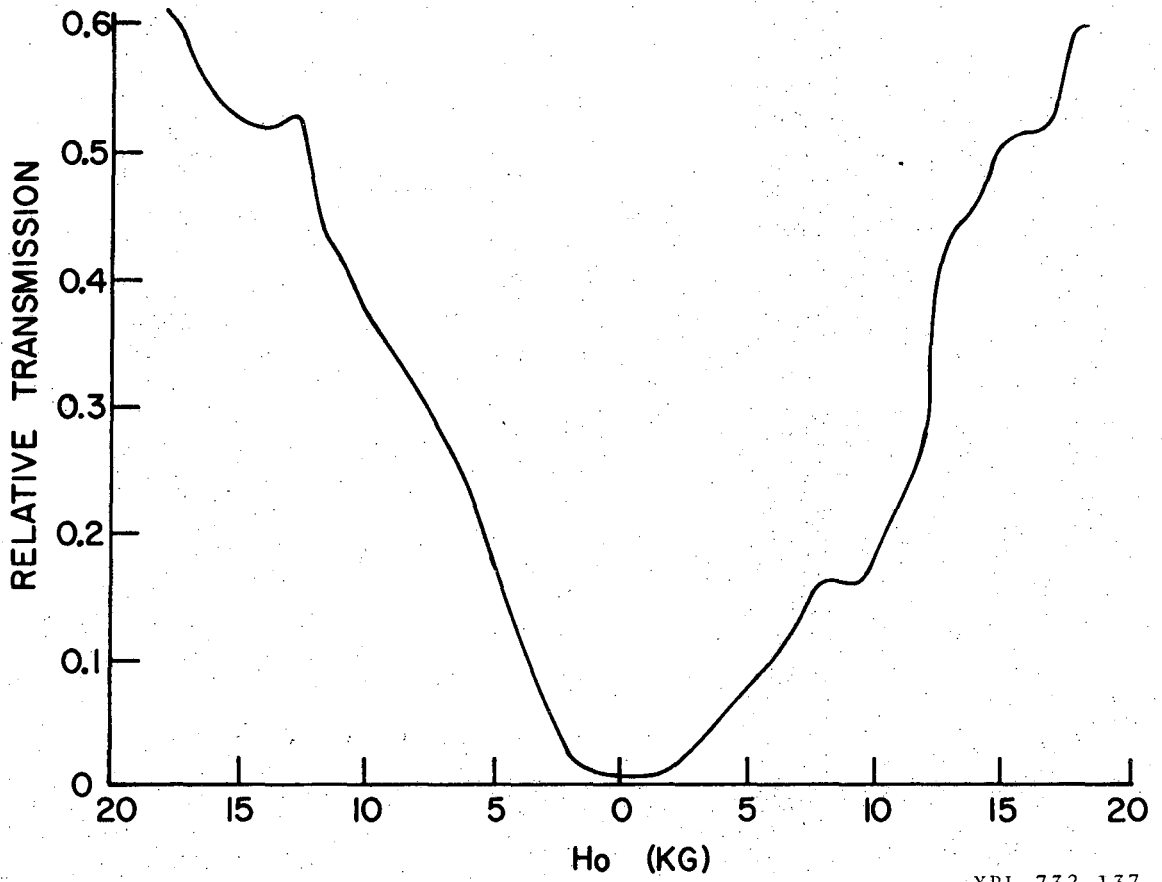
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Fig. 2.



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Fig. 3.



XBL 732-137

Fig. 4a.

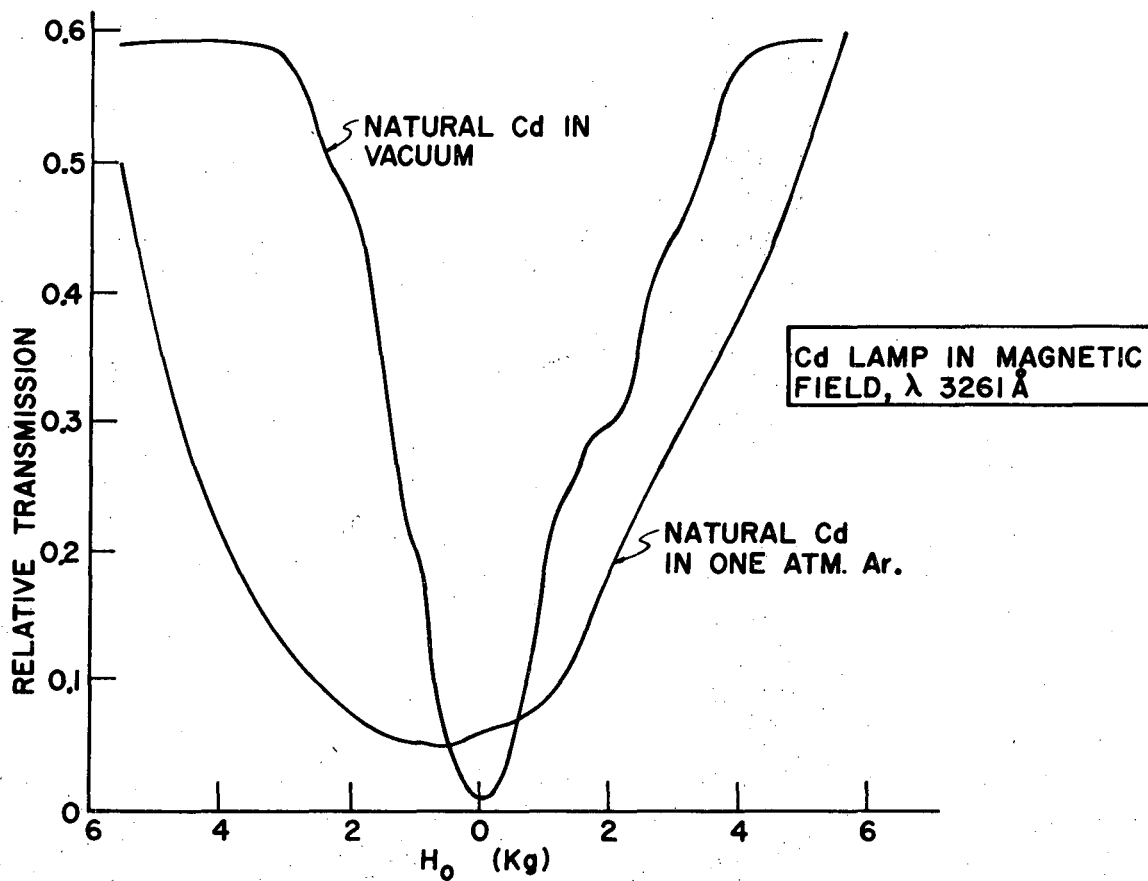
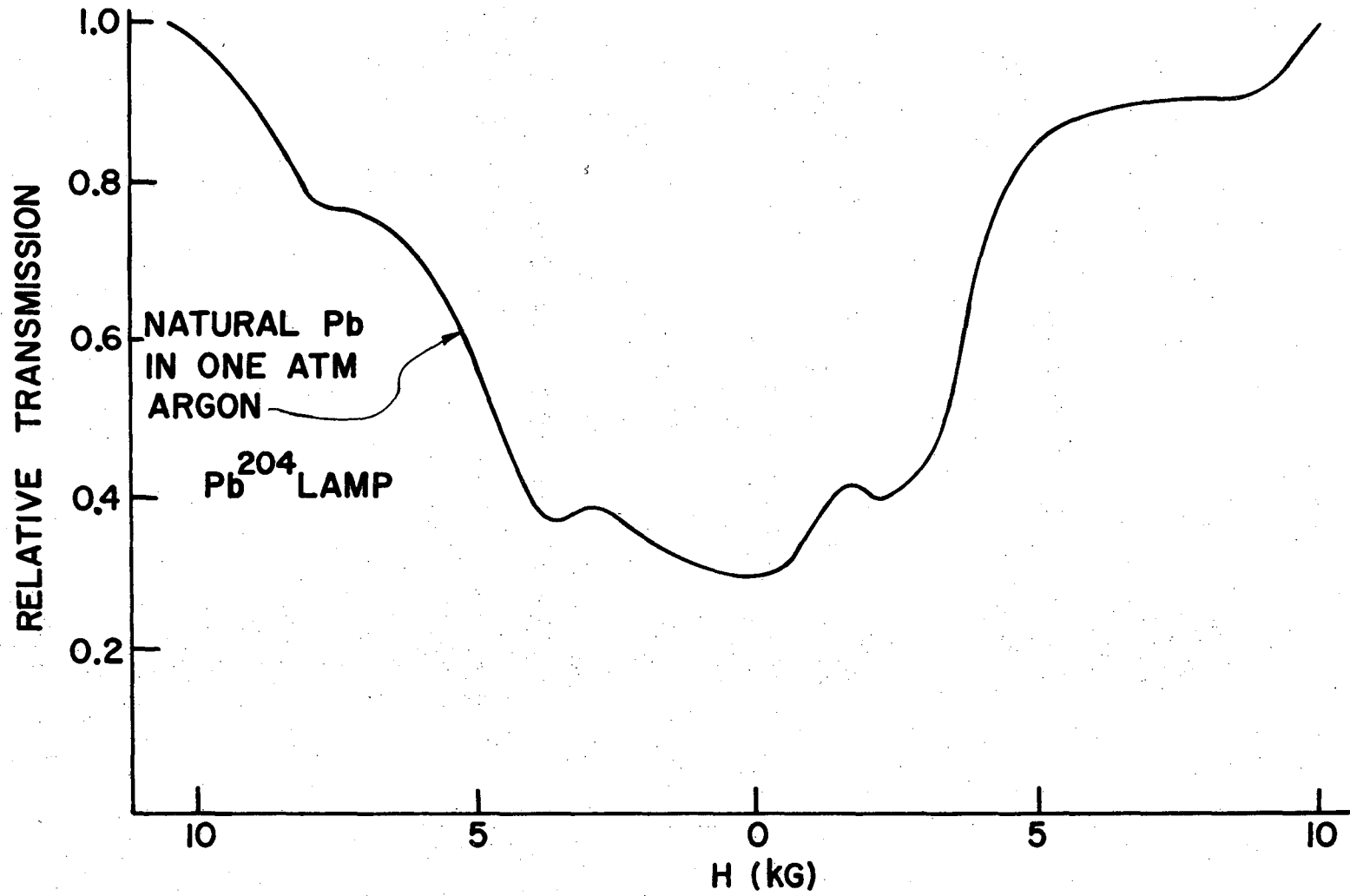


Fig. 4b.



XBL 7312-6837

Fig. 4c.

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