

Polarization of Light Resulting from the Excitation of Helium by Electrons*

ROBERT H. MCFARLAND AND EDWARD A. SOLTYSIK

Lawrence Radiation Laboratory, University of California, Livermore, California

(Received March 28, 1962)

The polarization of light resulting from the excitation of helium by electrons was measured as a function of the bombarding electron energy. The wavelengths of the light whose polarization was measured are as follows: $\lambda=4713 \text{ \AA}$ ($4^3S \rightarrow 2^3P$), $\lambda=4922 \text{ \AA}$ ($4^1D \rightarrow 2^1P$), $\lambda=3188 \text{ \AA}$ ($4^3P \rightarrow 2^3S$), $\lambda=5876 \text{ \AA}$ ($3^3D \rightarrow 2^3P$), $\lambda=4388 \text{ \AA}$ ($5^1D \rightarrow 2^1P$), $\lambda=5016 \text{ \AA}$ ($3^1P \rightarrow 2^1S$), $\lambda=4471 \text{ \AA}$ ($4^3D \rightarrow 2^3P$), $\lambda=3889 \text{ \AA}$ ($3^3P \rightarrow 2^3S$). The polarization curves for the different wavelengths were measured for different pressures of the helium gas, and for $\lambda=4922 \text{ \AA}$, 4388 \AA , and 5016 \AA , there is a marked pressure dependence. There is a decided discrepancy between the theory as summarized by Percival and Seaton and the experimental results presented in this paper. No resolution of this discrepancy is given at this time.

INTRODUCTION

THE experimental work of Skinner¹ and Skinner and Appleyard² on the polarization of light emitted by mercury when excited by electrons, gave results which were not in agreement with the theory of Oppenheimer³ and Penny.⁴ Lamb and Maiman⁵ measured the polarization of the 3889 \AA helium line and found an anomalous dependence of the polarization on the energy of the bombarding electrons.

A recent discussion of the theory of the polarization of light resulting from the excitation of atoms by electrons has been presented by Percival and Seaton.⁶ Included in their paper was a comparison between theory and the limited experimental data which were then in existence. They indicated that there is reasonable agreement between experiment and theory at electron energies greater than a few volts above threshold. Also, extrapolation of the higher energy polarization curves toward threshold energies generally led to a threshold polarization value in agreement with theory. In direct conflict, however, were the measured threshold values with those determined theoretically. With the possible exception of the He 3889 \AA , all measured polarization

percentages have uniformly approached zero as the electron energy approached the threshold for excitation.

Many authors have discussed atomic scattering and excitation processes, with the review article of Gerjuoy⁷ being among the more recent. Common among their observations is the importance that must be attached to atomic scattering processes in establishing and maintaining high-temperature plasmas. Equally common is the observation of the discrepancy between theory and measurement of low-energy excitation phenomena particularly as it concerns the threshold region.

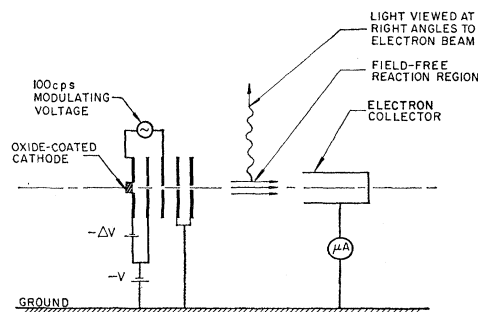


FIG. 2. Electron-gun electrode arrangement.

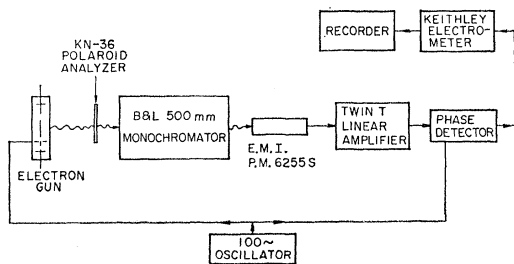


FIG. 1. Block diagram of experimental arrangement.

* This work was done under the auspices of the U. S. Atomic Energy Commission.

¹ H. W. B. Skinner, Proc. Roy. Soc. (London) **A112**, 642 (1926).

² H. W. B. Skinner and E. T. S. Appleyard, Proc. Roy. Soc. (London) **A117**, 224 (1927).

³ J. R. Oppenheimer, Z. Physik **43**, 27 (1927); Proc. Natl. Acad. Sci. U. S. **13**, 800 (1927).

⁴ W. G. Penny, Proc. Natl. Acad. Sci. U. S. **18**, 231 (1932).

⁵ W. E. Lamb and T. H. Maiman, Phys. Rev. **105**, 573 (1957).

⁶ I. C. Percival and M. J. Seaton, Phil. Trans. Roy. Soc. London **113**, 251 (1958).

⁷ E. Gerjuoy, Revs. Modern Phys. **33**, 544 (1961).

⁸ The polarization P is defined as $P = [(I_{II} - I_{\perp}) / (I_{II} + I_{\perp})] \times 100$, where I_{II} is the intensity of the radiation measured perpendicular to the electron beam, whose electric vector is parallel to the electron beam, and I_{\perp} is the intensity whose electric vector is perpendicular to the electron beam.

⁹ R. H. McFarland and E. A. Soltysik, Lawrence Radiation Laboratory Report UCRL-6749, 1962 (unpublished). (This is the complete report of which the present paper is a condensation.)

¹⁰ R. H. McFarland and E. A. Soltysik, Bull. Am. Phys. Soc. **6**, 423 (1961).

TABLE I. Polarization formulas.^a

λ (Å)	Transition	Theoretical polarization formula (references 6 & 9)	Polarization at threshold (%)
4388	$5^1D_2 \rightarrow 2^1P_1$	$P = 3 \left(\frac{\sigma_0 + \sigma_1 - 2\sigma_2}{5\sigma_0 + 9\sigma_1 + 6\sigma_2} \right) \times 100$	60
4922	$4^1D_2 \rightarrow 2^1P_1$	(Same as above)	60
5015	$3^1P_1 \rightarrow 2^1S_0$	$P = \left(\frac{\sigma_0 - \sigma_1}{\sigma_0 + \sigma_1} \right) \times 100$	100
3889	$3^3P_{2,1,0} \rightarrow 2^3S_1$	$P = 15 \left(\frac{\sigma_0 - \sigma_1}{41\sigma_0 + 67\sigma_1} \right) \times 100$	36.6
3188	$4^3P_{2,1,0} \rightarrow 2^3S_1$	(Same as above)	36.6
4471	$4^3D_{3,2,1} \rightarrow 2^3P_{2,1,0}$	$P = 213 \left(\frac{\sigma_0 + \sigma_1 - 2\sigma_2}{671\sigma_0 + 1271\sigma_1 + 1058\sigma_2} \right) \times 100$	31.7
5876	$3^3D_{3,2,1} \rightarrow 2^3P_{2,1,0}$	(Same as above)	31.7
4713	$4^3S_1 \rightarrow 2^3P_{2,1,0}$	$P = 0$	0

^a This table gives the polarization formulas for the lines investigated. σ_m is the cross section to an atomic state with magnetic quantum number m .

EXPERIMENTAL APPARATUS AND METHOD

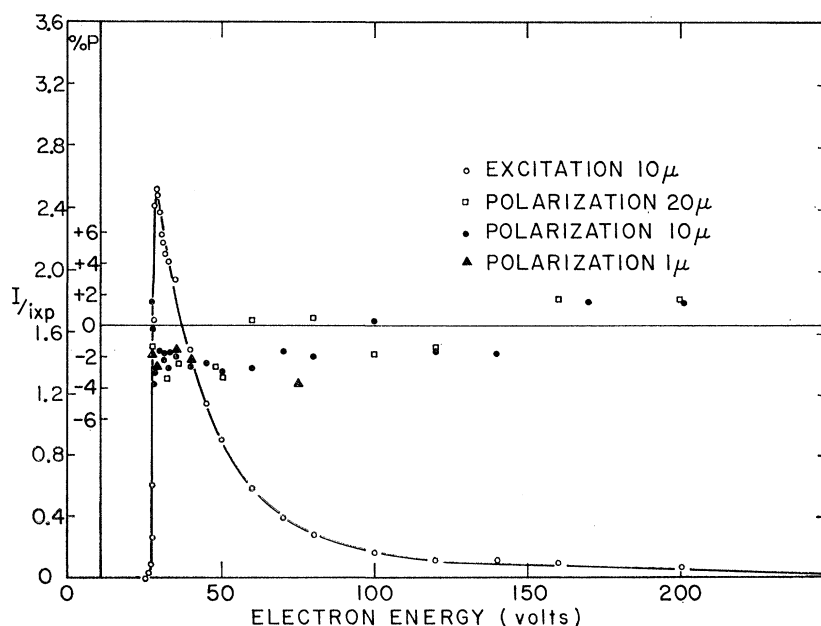
The apparatus used for this investigation is shown schematically in Fig. 1. The modulated light signal originating in the electron gun chamber passed through a quartz window, a Polaroid analyzer, a monochromator, and a photomultiplier tube. The resulting electrical signal was amplified with a narrow-band, lock-in, 100-cycle amplifier and was then phase detected. The dc output voltage drove a 10-mV recorder through an electrometer used as a voltmeter.

Figure 2 indicates schematically the plan of the excitation region of the system. The electron beam originated at an indirectly heated oxide-coated nickel cathode. Acceleration occurred through four disk-

shaped stainless-steel electrodes (coated with Aquadag) spaced 1 mm apart. The apertures limiting the beam were 30 mils in diam. With an axial magnetic field of 10 G, it was possible to account for all of the electrons emitted by the cathode as being collected by the accelerating electrodes and the electron collector located approximately 2 in. from the final accelerating electrode.

During a run, the cathode was set at a negative 15 V with respect to the first accelerating electrode whose potential was varied to supply the appropriate accelerating potential, $V + \Delta V$. Two Fluke model 407 power supplies and a Fluke model 801 HR differential voltmeter were used to provide and measure these accelerating voltages. The modulating voltage to the second

FIG. 3. Polarization and excitation vs energy. $\lambda = 4713\text{Å}$ ($4^3S \rightarrow 2^3P$).



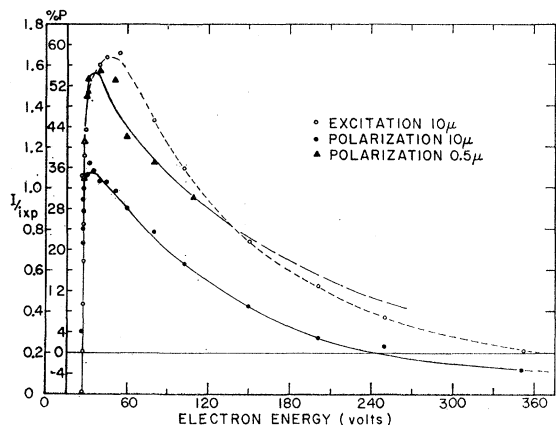


FIG. 4. Polarization and excitation vs energy.
 $\lambda = 4922\text{\AA}$ ($4^1D \rightarrow 2^1P$).

electrode was provided by a Hewlett-Packard model 200 CD audio oscillator. This voltage had no or at most a minimal effect on the final energy of the electrons reaching the reaction region but did block the beam one hundred times per second to provide a modulated light signal.

A Fluke model 408A power supply was used to provide the required voltage to the photomultiplier. Because of the 100-cps ac light signal and the rejection of all dc signals by the narrow-band twin T amplifier and phase-sensitive detector, it was unnecessary to cool the photomultiplier tube. The amplifier possessed a 10^5 amplification factor, which made necessary the attenuation of all but the weaker signals from the photomultiplier.

A built-in time constant of the order of 10 sec was used in the output of the phase detector for the weaker

signals. A visual averaging of the recorder response to a signal allowed for a more accurate measurement than was possible with the electrometer alone.

The electron gun and reaction region were connected to a 304 stainless-steel vacuum system pumped by a Varian 3-liter Vacorb pump and an 8-liter/sec Vacion pump. Copper pinch gaskets and Granville-Phillips 1-in. copper seat valves were used in the high-vacuum section of the system. After baking and before filling, a 10^{-8} Torr vacuum was obtainable. (1 Torr = 1 mm Hg.) Helium was admitted to the system through an auxiliary vacuum section consisting of a Welsch Duoseal pump and a liquid-nitrogen-cooled copper trap. This section was capable of 10^{-5} Torr, while the high-vacuum section pumped only with a Vacorb pump was capable of 10^{-6} Torr or less.

Experimental-grade helium was introduced slowly into the reaction region through the liquid-nitrogen-cooled copper trap. The Vacorb pump was open to the reaction region during filling and during the experiment. Experience indicated that CO, CO₂, and possibly hydrocarbons and water created in the electron gun and reaction chamber, as well as residual atmospheric gases, were continuously pumped by the Vacorb pump. This added to the helium purity and appeared to increase the required threshold potentials by approximately 2 V over those observed when these impurities were allowed to remain in the system. Whether the pump was used or not made little difference in the character of the polarization and excitation observed relative to threshold.

Pressure measurements were made with ionization and thermocouple gauges periodically calibrated for helium against a McLeod gauge attached to the system.

Instrumentally induced polarization errors were eliminated from the observations at each wavelength

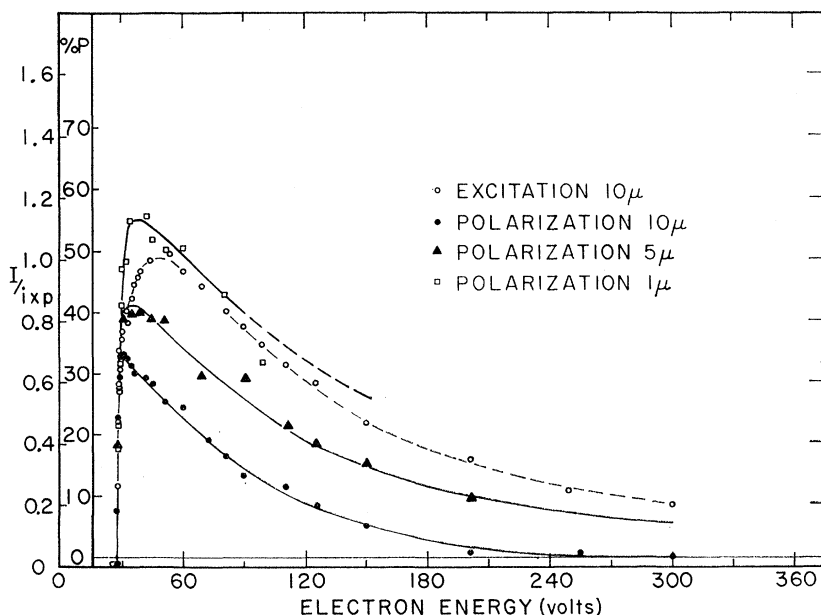


FIG. 5. Polarization and excitation vs energy. $\lambda = 4388\text{\AA}$ ($5^1D \rightarrow 2^1P$).

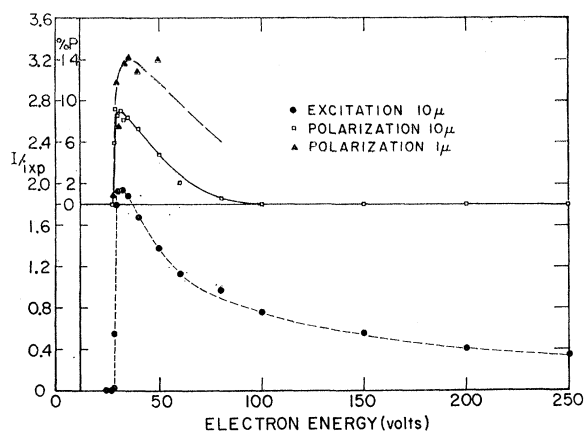


FIG. 6. Polarization and excitation vs energy.
 $\lambda = 4471\text{\AA}$ ($4^3D \rightarrow 2^3P$).

by comparison of the helium polarization with that of a mechanically chopped, unpolarized, white light source.

The half-intensity breadth of the electron beam under vacuum conditions was determined by retarding potential measurements to be 1 V. With helium in the gun the threshold of excitation was shifted to higher voltages. This was assumed to be due to contact potentials, and both the polarization and excitation curves were plotted together in order to establish the polarization relative to threshold.

EXPERIMENTAL RESULTS

Figures 3 through 13 show the energy dependence of excitation and polarization on bombarding electron energy. In each of these, the excitation curve is plotted

in relative units of intensity per microampere and per micron pressure. Because of the general linear dependence of excitation on pressure as determined by supporting experiments, only the 10- μ excitation measurements have been plotted. (Only helium 5016 \AA showed marked departure from a linear pressure dependence in agreement with work of St. John, Bronco, and Fowler.¹¹) On the other hand, polarization curves have been plotted for a few pressures to indicate the increase of polarization with decreasing pressure. Measurements were made of the polarization as a function of helium pressure at a fixed electron energy. For triplet-state transitions the polarization was independent of pressure below 5 μ , while for singlet-state transitions the polarization was pressure dependent above 0.5 μ . For singlet-state transitions the polarization at 5 μ was about 10% lower than at 1 μ .

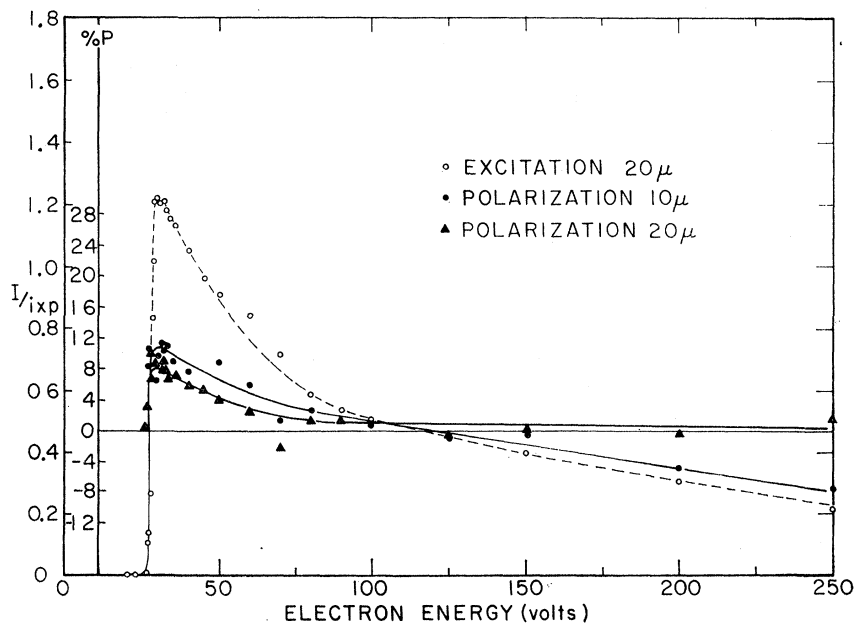
Additional measurements have been made to indicate that the polarization at a given pressure is not dependent upon electron beam current within the ranges used or upon axial magnetic fields up to the 10-G field used. Preliminary measurements have indicated that line shape does not change as one increases the electron energy within the range of this experiment, which implies that the radiation is not the result of an ion complex at the lower energies.

An additional observation which does not specifically involve polarization was that transitions corresponding to a sharp singlet series were not observed in the helium spectrum at energies near threshold.

DISCUSSION OF RESULTS

All of the excitation and polarization curves with the exception of those of helium 3889 \AA have features in

Fig. 7. Polarization and excitation vs energy. $\lambda = 5876\text{\AA}$ ($3^3D \rightarrow 2^3P$).



¹¹ R. M. St. John, C. J. Bronco, and R. G. Fowler, J. Opt. Soc. Am. **50**, 28 (1960).

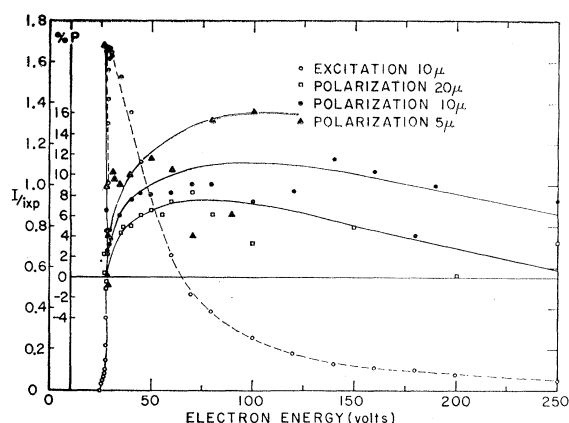


FIG. 8. Polarization and excitation vs energy.
 $\lambda = 3188\text{\AA}$ ($4^3P \rightarrow 2^3S$).

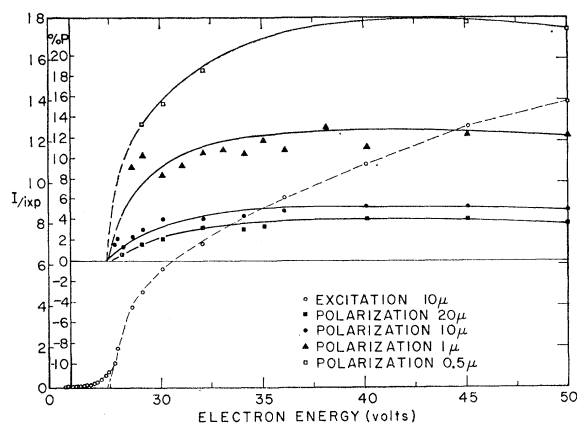


FIG. 9. Polarization and excitation vs energy.
 $\lambda = 5016\text{\AA}$ ($3^1P \rightarrow 2^1S$).

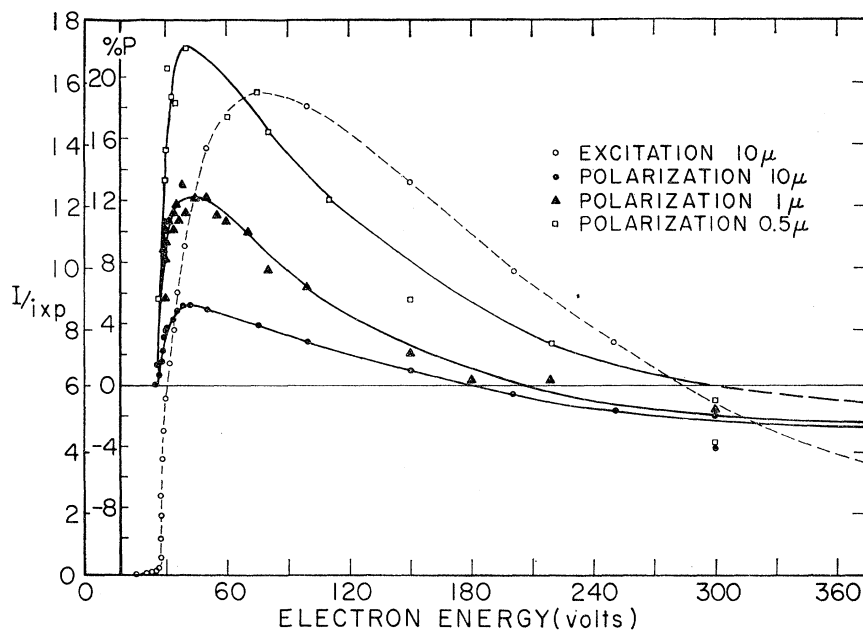


FIG. 10. Polarization and excitation vs energy. $\lambda = 5016\text{\AA}$ ($3^1P \rightarrow 2^1S$).

common. Polarization is observed to increase with decreasing pressure. At electron energies of 20 V or so above threshold, polarization curves at low pressures (notably 4388 and 4922) conform to "expected" theoretical values. Extrapolation of these curves to threshold results in values approximating theory, as has been observed for mercury.⁶

However, measured polarization curves, with the possible exception of 3889 Å, have been observed to have zero polarization at threshold and to increase rapidly to maximal values at 10 to 20 V above threshold. The disagreement at threshold between theory (see Table I) and the observations reported here is puzzling because the theoretical prediction at threshold depends simply on the assumption of Russell-Saunders coupling and the conservation of orbital angular momentum. It would appear that the experimental results can only be understood providing one relinquishes the assumption that both L , the total orbital angular momentum, and S , the total spin, are conserved separately. Baranger and Gerjuoy¹² indicated that a "compound ion" model of the phenomenon, as opposed to a direct excitation process as envisaged here, might make the "apparent" nonconservation of L and S more believable.

Because the percent of polarization as measured is defined as

$$P = [(I_{\parallel} - I_{\perp}) / (I_{\parallel} + I_{\perp})] \times 100,$$

it is interesting to discuss the low-pressure polarization of a line such as 4388 Å, Fig. 5, in terms of I_{\parallel} and I_{\perp} . Their variation on an expanded voltage scale appears in Fig. 14.

¹² E. Baranger and E. Gerjuoy, Proc. Phys. Soc. (London) **72**, 328 (1958).

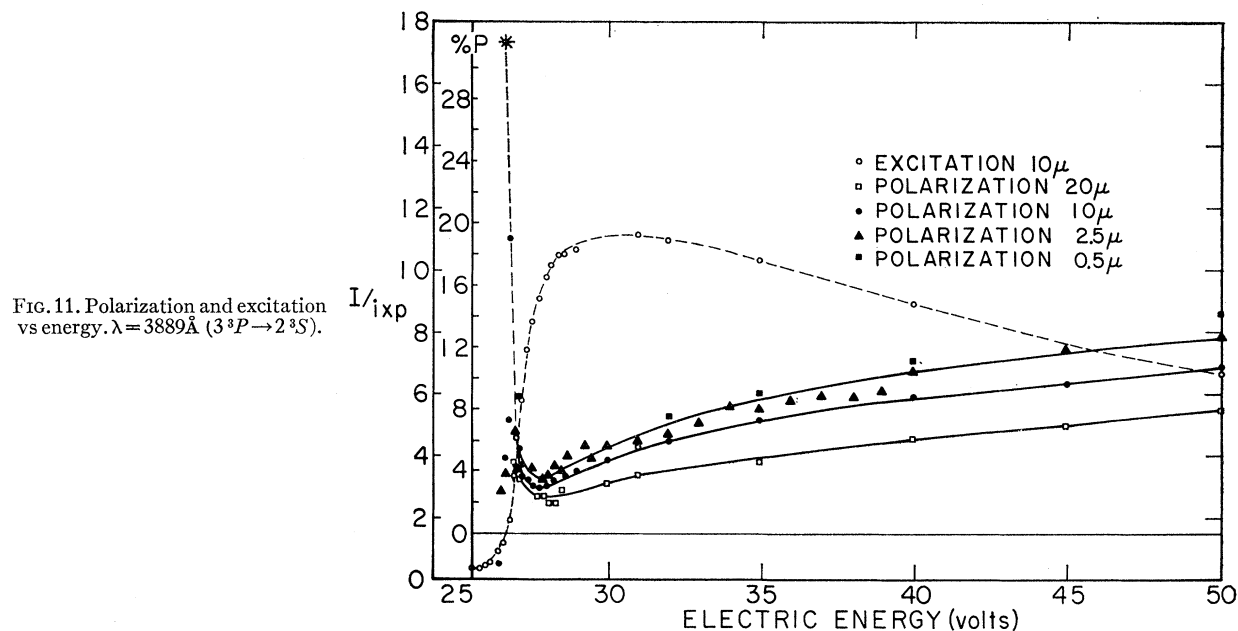


FIG. 11. Polarization and excitation vs energy. $\lambda = 3889\text{\AA}$ ($3^3P \rightarrow 2^3S$).

At electron energies 12 V above threshold the polarization is a maximum and P , I_{11} , I_1 , $(I_{11} + I_1)$, and $(I_{11} - I_1)$ may be considered as linear decreasing functions at least to 100 V electron energy. Between a correct threshold energy of 24 V and an electron energy of 36 V, each of these functions becomes less well known. However, $I_{11} = I_1 = 0$ at threshold. Attempts to correlate these intensities near threshold E_T with the $E(E - E_T)^{1/2}$ law have been unsuccessful as has the attempt to correlate this square root law with $(I_{11} + I_1)$. Rather, the exci-

tation within a few tenths of a volt of threshold appears to follow a linear dependence of $(E - E_T)$ as was observed in a different type of experiment by Schultz.¹³ For the extent of this linear dependence, the polarization must of necessity be constant and small as the increasing slopes of the I_{11} and I_1 curves are large and nearly equal. Above $E - E_T = 1/2$ V, I_{11} continues to rise while I_1 remains more nearly constant. Even at this low pressure, the intensity of I_{11} is great enough at a few tenths volt above threshold that its accuracy is of the order of

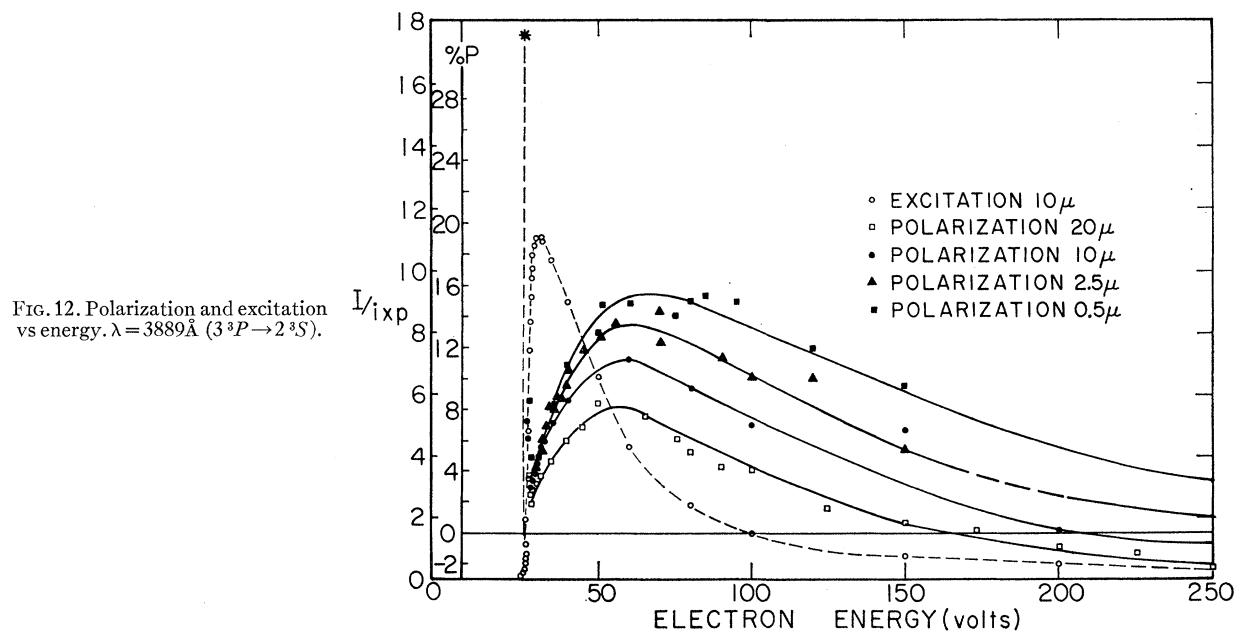


FIG. 12. Polarization and excitation vs energy. $\lambda = 3889\text{\AA}$ ($3^3P \rightarrow 2^3S$).

¹³ G. J. Schultz, Phys. Rev. **112**, 150 (1958).

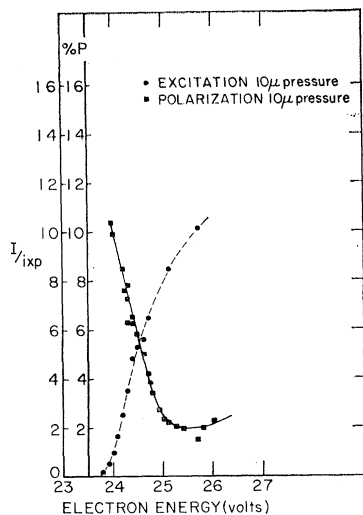


FIG. 13. Excitation and polarization vs energy near threshold. $\lambda = 3889\text{\AA}$ ($3^3P_{2,1,0} \rightarrow 2^3S_1$).

1 or 2%. Assuming the I_{II} curve drawn is correct and the polarization extrapolation to the theoretical polarization threshold as being linear, one may calculate a necessary I_I curve. This is shown as the dashed curve which intercepts the I_I curve at approximately 40 V. Except for the high measurements close to threshold (first 5 V), measured values conform within experimental error to calculated values. Near threshold, however, measured values are as great as three times the needed calculated ones. These differences are considered to be greater than experimental error.

The other striking anomaly observed in this work

involves the energy dependence of the polarization of 3889\AA , Figs. 11–13. No convincing explanation of this is available at this time although one may conjecture concerning resonance radiation absorption, transfer collisions, and cascade population along the lines taken by St. John and Fowler.¹⁴

The polarization of 4713\AA corresponding to a $4^3S \rightarrow 2^3P$ transition should have zero polarization at all energies. The negative 2% observed somewhat stretches the accuracy of the measurements at the intensities observed which are estimated at $\pm 1\%$. The apparent independence of pressure and electron energy would tend to support the feeling that the measured values should indeed be zero.

For the remaining lines, including 5016\AA , the maximum measured polarization falls far short of the predicted values. All lines appear to approach zero polarization at threshold. Helium 4471\AA , 5876\AA , and 3188\AA all suffer from low intensity so that low-pressure measurements were difficult. One might assume, however, that they would approach the appearance in shape of the 4388\AA and 4922\AA lines at a sufficiently low pressure.

CONCLUSIONS

Neither excitation nor polarization functions of energy as measured in this experiment approach the threshold as theoretically predicted.

The polarization observed for 3889\AA as a function of energy has two extremum points for which there are no convincing reasons.

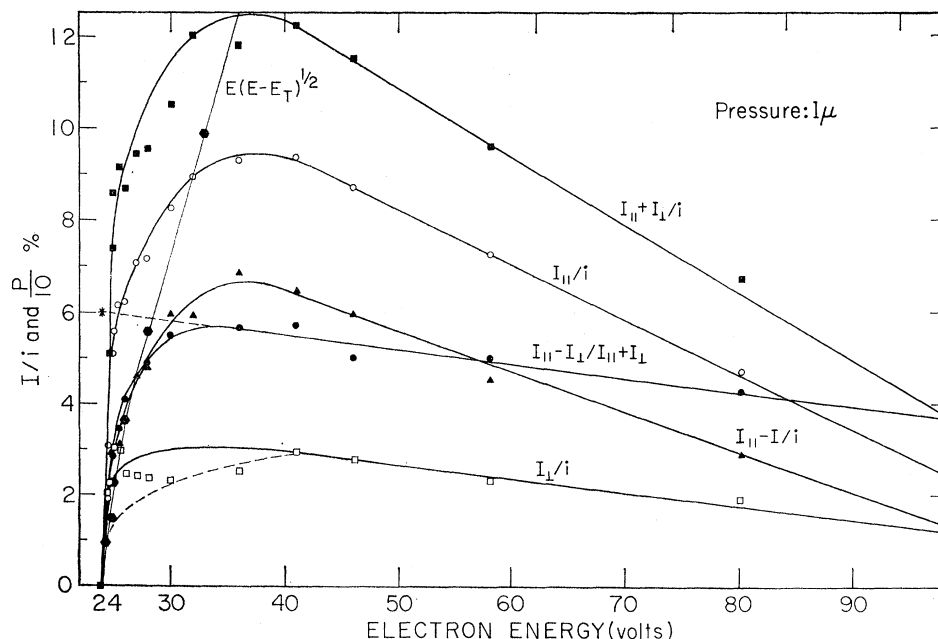


FIG. 14. Polarization and excitation vs energy. $\lambda = 4388\text{\AA}$ ($5^1D \rightarrow 2^1P$).

¹⁴ R. M. St. John and R. G. Fowler, Phys. Rev. **122**, 1813 (1961).

The present work tends to confirm the work of other experimentalists^{2,5,15,16} and adds to the list of spectral lines for which polarization measurements have been made. It emphasizes a lack of knowledge concerning one phase of atom scattering important in plasma production.

¹⁵ D. W. O. Heddle and C. B. Lucas, Abstracts, Second International Conference on the Physics of Electronic and Atomic Collisions, University of Colorado, (W. A. Benjamin, Inc., New York, 1961), p. 119.

¹⁶ R. H. Hughes, R. B. Kay, and L. D. Weaver, *14th Annual Gaseous Electronics Conference, 1961, General Electric Company, Schenectady, New York* [Bull. Am. Phys. Soc. **7**, 130 (1962)].

ACKNOWLEDGMENTS

The authors wish to thank Dr. C. M. Van Atta for lending his support to this work. Professor B. Curnutte and W. Tobey of Kansas State University, during their visit to the Lawrence Radiation Laboratory in the summer months of 1961, contributed their efforts to this work's advancement. Also, the authors wish to thank C. Alexander, J. Fabyan, R. Copp, and other members of the Atomic Physics Group, and L. Ducote for assistance in the construction and maintenance of the experimental equipment.

Mössbauer Effect in Eu^{151} ; Possible Influence of Optical Branches*

D. A. SHIRLEY, M. KAPLAN, R. W. GRANT, AND D. A. KELLER

Lawrence Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California

(Received May 7, 1962)

The Mössbauer effect has been observed in the 21.7-keV transition in Eu^{151} . The absorption pattern is a single line, independent of temperature, as expected for an ion in which the ground level has $J=0$. A lower limit of 0.92×10^{-8} sec can be set on the lifetime of the isomeric state.

The 21.7-keV transition is, thus, about a factor of 100 slower than the single-particle estimate. The large discrepancy between the Debye temperatures required to fit the data of 77 and 295°K is attributed to the influence of optical branches.

I. INTRODUCTION

SOON after the initial discovery of recoil-free γ -ray resonance by Mössbauer in 1958,¹ the effect was observed in several nuclei. With the experimental realization of the very sharp 14.4-keV resonance in Fe^{57} ,² and particularly after the discovery of the nuclear Zeeman effect, it became clear that very few nuclei were qualified for detailed investigations of local fields. In fact the combined requirements of a reasonable lifetime for the isomeric state, an appropriate transition energy, and a low conversion coefficient, as well as other technical factors, have excluded all the Mössbauer nuclei save Fe^{57} and Sn^{119} from extensive application. While these two nuclei have been used in a great variety of chemical environments, there are many potential problems for which they are not suitable. In particular, it was desirable to find a Mössbauer resonance in an element having the chemical properties of a rare earth. With this goal the present research was undertaken.

In this paper we discuss the spectroscopy associated with the 21.7-keV resonance in Eu^{151} , and some experimental problems, as well as reporting the resonance and

some rather tentative quantitative results obtained with oxide sources and absorbers.

II. ELECTRONIC SPECTROSCOPY OF TRIVALENT EUROPIUM

Rare-earth ions have electronic configurations of the form $4f^n$. In Russell-Saunders coupling these form terms

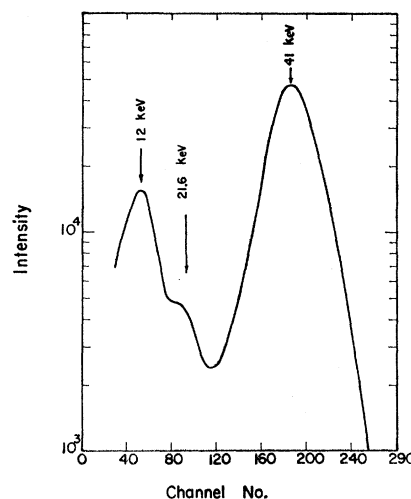


FIG. 1. Low-energy gamma-ray spectrum of Gd^{151} obtained with a $11/2 \times 1/4$ -in. $\text{NaI}(\text{Tl})$ scintillation counter. The 12-keV peak is the K x-ray escape peak; the 21.7-keV peak is the photo peak; the 41-keV peak is the K x ray.

* Work done under the auspices of the U. S. Atomic Energy Commission.

¹ R. L. Mössbauer, *Naturwissenschaften* **45**, 538 (1958); *Z. Physik* **151**, 125 (1959); *Z. Naturforsch.* **14a**, 211 (1959).

² R. V. Pound and G. A. Rebka, Jr., *Phys. Rev. Letters* **3**, 554 (1959). J. P. Schiffer and W. Marshall, *ibid.* **3**, 556 (1959).