provide a useful tool to improve color reproduction still further.

ACKNOWLEDGMENTS

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Some Quantitative Aspects of an Opponent-Colors Theory. I. Chromatic Responses and Spectral Saturation

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Saturation discrimination is assumed to be dependent on the ratio of chromatic to achromatic components in the sensory response to a given wavelength. The usual methods of measurement do not, however, permit independent control of the chromatic and achromatic variables. On the basis of an opponent-colors theory of vision, a method is described for measuring directly and separately the spectral distributions of the chromatic components. A series of experiments is reported in which this method was used to obtain measures of the paired chromatic responses associated with the four primary spectral hues. Results are reported for two observers, 1° foveal test field, for an equal brightness spectrum (10 mL), and a neutral state of adaptation (10 mL). The measured chromatic responses, together with achromatic (luminosity) functions measured earlier for the same observers, are used to predict the form of the spectral saturation discrimination function.

INTRODUCTION

THE saturation of a color is related to the perceived amount of hue in the color sensation. It is easy to demonstrate that intermixing gradually increasing quantities of white light with pure spectral colors makes the latter increasingly pale until eventually they become colorless. Under such circumstances they are said to become less saturated. However, even without the intermixture of white light, it is quite obvious from only a cursory examination of the spectrum that the various spectral regions are intrinsically unequally saturated.[1] Furthermore, with a large increase in stimulus radiance saturation is modified so that eventually the spectral hues are seen as white.[2]

Experimentally, a number of different techniques have been used to determine the functional dependencies between saturation and the associated physical variables, wavelength and radiance. Most of the data on spectral saturation in relation to wavelength have been obtained by measuring changes in colorimetric purity to produce just perceptible changes in saturation. This method assumes that the threshold change in colorimetric purity varies inversely with the ratio of chromatic to achromatic excitations in the visual response to a stimulus of given wavelength. The minimal perceptible change in colorimetric purity has been measured in two ways, either by adding monochromatic to white light or by adding white light to monochromatic. Data obtained by adding spectral lights to white have been reported by a number of investigators including Purdy; Priest and Brickwedde; Martin, Warburton, and Morgan; Wright and Pitt; Nelson; Grether; and Chapanis.[3–9] The results are generally similar: the spectral saturation function shows maxima in the short and long wave regions with a sharp minimum in the 560 to 580-mμ region.

Other experimenters have employed other measures of saturation, all of which are assumed to be either directly or inversely related to the percentage of hue in the color response. For a series of spectral colors, Jones and Lowry stepped off in just noticeable difference steps the distance between each spectral color and white[10]; Troland measured the rate at which flicker ceases when a chromatic stimulus is alternated with white in the flicker photometer[11]; and Sinden measured the "complementation valence" of spectral lights, i.e., the luminosity ratios of the two monochromatic components

[1] L. T. Troland, *The Principles of Psychophysiology* (D. Van Nostrand Company, Inc., New York, 1930), Vol. II, p. 147.
[2] J. H. Parsons, *An Introduction to the Study of Colour Vision* (Cambridge University Press, Cambridge, England, 1924), pp. 31–32.
[3] D. M. Purdy, "Chroma as a function of retinal illumination," dissertation, Harvard University, 1–190 (1929).
[4] I. G. Priest and F. G. Brickwedde, J. Opt. Soc. Am. 28, 133 (1938).
[5] Martin, Warburton, and Morgan, Med. Research Council (Brit.), Spec. Rept. Ser. No. 188, (1933).
[6] W. D. Wright and F. H. G. Pitt, Proc. Phys. Soc. (London) 49, 329 (1937).
[7] J. H. Nelson, Proc. Phys. Soc. (London) 49, 332 (1937).
[8] W. F. Grether, J. Exptl. Psychol. 28, 419 (1941).
[9] A. Chapanis, J. Exptl. Psychol. 34, 24 (1944).
[10] L. A. Jones and E. M. Lowry, J. Opt. Soc. Am. 13, 25 (1926).
[11] L. T. Troland, Trans. Illum. Eng. Soc. (N. Y.) 11, 947 (1916).

in complementary light pairs.[12] The functions resulting from these different experimental procedures again show a general similarity and they approximate those obtained by measuring the minimal perceptible change in colorimetric purity when spectral light is added to white.

Still two more saturation indices have been reported by Purdy.[13] In one, the values of the luminance range between the absolute light threshold and the color threshold (achromatic interval, photochromatic interval) were measured as a function of wavelength, in the other, he determined the luminance required for maximal saturation as a function of wavelength. In the latter instance the optimal luminance for saturation is found to be smallest at the spectral extremes and greatest in the yellow region (565 mμ), whereas a graphical representation of the achromatic interval against wavelength shows only a very rough approximation to this recurring pattern.

There is a single exception to the general uniformity of functions obtained by different measures of spectral saturation. The single exception concerns experiments where the minimal perceptible change in colorimetric purity is measured by adding white light to monochromatic light. In terms of the underlying assumption, it was expected that it would be irrelevant whether a saturation difference is produced by adding white light to spectral light or spectral light to white. As we have noted, when spectral light is added to white there results the commonly recognized saturation function. On the other hand, in the reverse procedure, i.e., white light added to monochromatic, the function as measured by Wright and Pitt[14] and as derivable from the Jones and Lowry data,[10] is nearly constant and independent of wavelength. In seeking to account for this unanticipated result Wright and Pitt suggest that differences in initial states of adaptation might account for the different results in the two procedures. This specific "anomalous" result will be discussed in a later paper of this series.

Theoretical discussions of the spectral saturation function are very limited. Wright summarizes the weakness of the classical three-component views with regard to this function when he says "in some ways the data represented by this diagram . . . are among the most difficult to explain on a three-response theory of the classical type. If the red, green, and blue radiations in the spectrum each stimulated only one of the three response systems, we might expect that the sensations they aroused would be approximately of equal saturation. Similarly, mixtures of red and green (yellow), blue and green (blue-green), and blue and red (purple) might be expected to be equally strongly saturated. Although it is certain that no spectral radiation does stimulate only one of the receptor processes, nevertheless most three-response theories assume that red, green, and blue spectral radiations give rise to responses which are dominated by the red, green, and blue receptor systems, respectively. It is therefore difficult to explain the marked desaturation of the 0.56–0.57-μ region of the spectrum. . . ."[15]

Nevertheless, since spectral mixture curves are based upon a more or less arbitrary selection of primaries it is, of course, quite legitimate to recompute them in such a way as to obtain the best possible quantitative fit for any specific visual function or psychophysical speculation. Both Judd[16] and Hecht[17] have done exactly this for the spectral saturation function. The two specific and different formulations of the Young-Helmholtz theory suggested by these investigators enable them to derive theoretical saturation functions which fit the Priest and Brickwedde experimental curves very closely. As Judd has indicated, however, specific mixture curves useful for interpreting specific functions frequently have serious shortcomings in respect of other visual properties. In this instance, for example, both explanations fail to account for the facts of dichromatic vision.[18]

A satisfactory quantitative account of the saturation function that is of more than very restricted validity is given by the Müller-Judd formulation based on the Hering opponent-colors theory.[18] In this formulation, the theoretical derivations involve transformations to three different sets of distribution functions that express not only the color-mixture relations but also the relations of the four primary hue responses. In this way Judd was able to account not only for the chromatic sensibility data but also for various dichromatic color systems. Since it has long been recognized that color mixture data may be adequately described in terms of any of an infinity of mathematically related sets of distribution functions, the mathematical validity of Judd's theoretical analysis is beyond question and Hecht's statement concerning the number of distribution functions is still true. "The problem is. . . essentially a physiological one, and not a mathematical one."[19]

An important aspect of this theoretical analysis is the separation of visual responses to color into their individual chromatic and achromatic components. An analytical separation of this sort, if valid, should lend itself to verification by psychophysical experimentation, and maximal information about saturation as well as other aspects of color sensations may be expected to come from such psychophysical experiments.

The experimental approach permitting differentiation of the various chromatic responses *per se* is implicit in the Hering opponent-colors theory.[20] A basic assumption

[12] R. H. Sinden, J. Opt. Soc. Am. **7**, 1123 (1923).
[13] D. M. Purdy, Brit. J. Psychol. **21**, 283 (1931).
[14] W. D. Wright and F. H. G. Pitt, Proc. Phys. Soc. (London) **47**, 205 (1935).
[15] W. D. Wright, *Researches on Normal and Defective Colour Vision* (C. V. Mosby Company, St. Louis, 1947), p. 165.
[16] D. B. Judd, Natl. Bur. Standards (U. S.) 4, 515 (1930).
[17] S. Hecht, *A Handbook of General Experimental Psychology* (edited by C. Murchison) (Clark University Press, Massachusetts, 1934), pp. 704–828.
[18] D. B. Judd, Natl. Bur. Standards (U. S.) **42**, 1 (1949).
[19] Reference 17, p. 792.
[20] E. Hering, Sitzber. Akad. Wiss. Wien Math.-naturw. Klasse. **70**, 169 (1875).

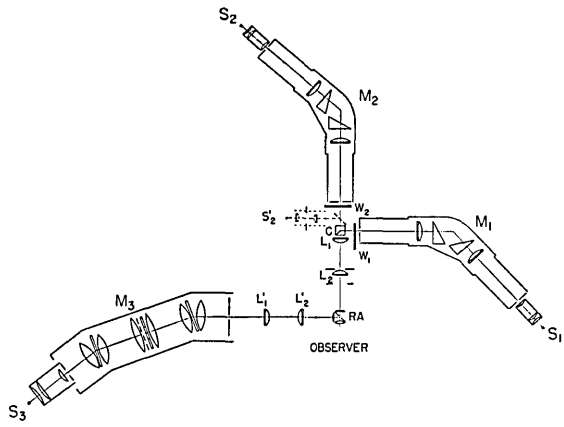

FIG. 1. Schematic diagram of optical system.

in this theory is the antagonism or the opponent nature of the two components within the paired yellow-blue and red-green visual processes. Thus, a yellow response of given magnitude should be exactly canceled by a stimulus that elicits the same magnitude of blue response, and a similar relation should hold between red and green responses. This view leads directly to an experimental approach in which a null method can be used to measure the spectral distributions of the separate chromatic responses. By relating the stimulus energy of a spectral wavelength evoking one hue to the stimulus energy of a wavelength that excites an opponent hue of exactly equivalent magnitude, the spectral distributions of the four primary hue responses can be directly determined. The feasibility of such an experimental approach has been indicated by the preliminary results obtained by Brückner in an investigation of this type.[21,22]

APPARATUS

The apparatus used in these experiments has been fully described elsewhere.[23] It comprises, in essence, two Farrand prism monochromators and a diffraction-grating double monchromator of zero dispersion. A schematic diagram of the optical system is shown in Fig. 1. For use in the present experiments the optical cube designated as C in Fig. 1 was replaced by a 50 percent reflector. This mirror bisected the 90° angle between the two monochromators M_1 and M_2 and permitted the physical mixture of the independently variable light stimuli from the two monchromators. Monochromator M_2 provided a series of 29 spectral test stimuli spaced at 10-mμ intervals and monochromator M_1 provided the four mixture stimuli. The superimposed test and mixture stimuli were seen fused in a 1°×0.8° slightly elliptical field, which was fixated foveally. The binary mixture field was surrounded by a 37° adapting field provided by monochromator M_3 and this field was adjusted to be a chromatically neutral white for each observer.[24] The radiant flux of each of the monochromators M_1 and M_2 was controlled by calibrated neutral density Wratten filters and wedges located at the exit slits W_1 and W_2 as indicated in Fig. 1. The energy and wavelength calibrations of the apparatus have been fully described in a previous paper.[23]

METHOD AND PROCEDURE

The experimental procedure used to determine the distributions and magnitudes of chromatic valences or chromatic responses evoked by spectral stimuli is based on the assumption that the yellow and blue and red and green chromatic systems are antagonistic or opponent pairs.[25,26] In the spectrum in which the hues normally range from a slightly yellowish-red at one end through reddish-yellow, yellow, yellow-green, green, blue-green, blue to violet (reddish-blue) at the other end, there are only four primary hue components, and consequently only four "equilibration" or "compensatory" spectral stimuli are required for the chromatic response measurements: a red to balance the greens, a yellow to balance the blues, a green to balance the reds, and a blue to cancel the yellows.

The chromatic response of the visual system for a given hue is assumed to be proportional to the amount of the opponent cancellation stimulus necessary to extinguish that hue. To measure the amount, say, of yellow chromatic response evoked by a spectral test stimulus perceived as yellow, whether pure yellow, red-yellow, or green-yellow, the experimenter adds to the test stimulus a variable amount of blue stimulus (e.g., 467 mμ) until the observer reports that the yellow hue of the test stimulus is exactly canceled. In other words, the observer's endpoint is a hue (or a neutral sensation) that is neither yellow nor blue. For the series of test stimuli seen initially as red-yellows (approximately 700–580 mμ), the end point is a whitish red. For a series of green-yellow test stimuli (approximately 580–500 mμ) the end point is, of course, a green. For a yellow spectral stimulus that is unique or pure the observer's end point is a neutral colorless sensation.

When measuring the amount of red chromatic valence elicited by long- or short-wave test stimuli, a spectral stimulus that evokes a green hue is used for equilibration. At the long-wave end of the spectrum the un-

[21] A. Brückner, Z. Sinnesphysiol. 58, 322 (1927).

[22] Two other experiments of a similar nature have been reported [L. Goldytsch, Z. Biol. 67, 35 (1917); E. Brücke and N. Inouye, Pflüger's Arch. ges. Physiol. 141, 573 (1911)]. In one, only a single chromatic response function (yellow) was measured, in the other, chromatic valence curves were determined for a color-blind observer.

[23] L. M. Hurvich and D. Jameson, J. Opt. Soc. Am. 43, 485 (1953).

[24] D. Jameson and L. M. Hurvich, J. Exptl. Psychol. 41, 455 (1951).

[25] The phrase "chromatic valences" as used here is not to be confused with Hering's "optical valences" which refer to stimulus properties or capacities relative to a single, fixed state of adaptation (reference 26). Nor should the phrase "chromatic response" be confused with the so-called "fundamental response curves" or receptor sensitivity curves.

[26] E. Hering, Pflüger's Arch. ges. Physiol. 57, 308 (1894).

canceled "hue remainder" is yellow, while at the short wavelengths (reddish-blue) the "hue remainder" i.e., the uncanceled hue seen when both red and green hues have been eliminated is blue.[27] The analysis is similar for the remaining two series of spectral stimuli which have either a blue hue component (400–500 mμ) or a green one (480–580 mμ).

The wavelengths of the cancellation stimuli used for observer *H* were 467 mμ, 490 mμ, 588 mμ, and 700 mμ and for observer *J*, 475 mμ, 500 mμ, 580 mμ, and 700 mμ. For each observer the first three stimuli evoke hues that are perceptually unique blue, green, and yellow, respectively while the fourth cancellation stimulus (700 mμ) is a slightly yellowish-red for both observers. Pure or unique red is normally extraspectral but the use of any monochromatic band that contains red can serve as an equilibration stimulus.[21] There is, in fact, nothing critical about the use of perceptually unique equilibration stimuli. Their use is favored only because it simplifies analysis.

All chromatic response measurements were for an equal brightness spectrum of 10 mL. The luminance of the 37° white surround was also fixed at 10 mL throughout the entire experimental series.

At the beginning of each experimental session a 10-minute period of light exclusion was followed by a 5-minute period of bright adaptation to the white surround. The surround remained present and adaptation was maintained throughout each experimental session. The observer was told what specific combination of test and cancellation hues the experimenter was about to use, e.g., blue cancellation primary and spectral yellows. Upon a signal from the experimenter, the observer moved his fixation from the white surround to the center of the 1° dark area in the center of the surround. A mixture of test and cancellation stimuli was exposed in the 1° test area for a few seconds by means of a squeeze-bulb type shutter (Packard-Ideal) and the luminance of the cancellation stimulus adjusted in accordance with the observer's reports of the hue of the mixture pair. When the subject signaled that his end point had been achieved the shutter was closed and the observer again fixated the white surround. The mixture was then re-exposed (approximately 0.5–1.0 sec.) and the observer signified either acceptance of his initial setting or the change in the mixture desired. The readjustments were continued and brief exposures of the test field were repeated until the observer reported that the mixture met the necessary end-point criterion.

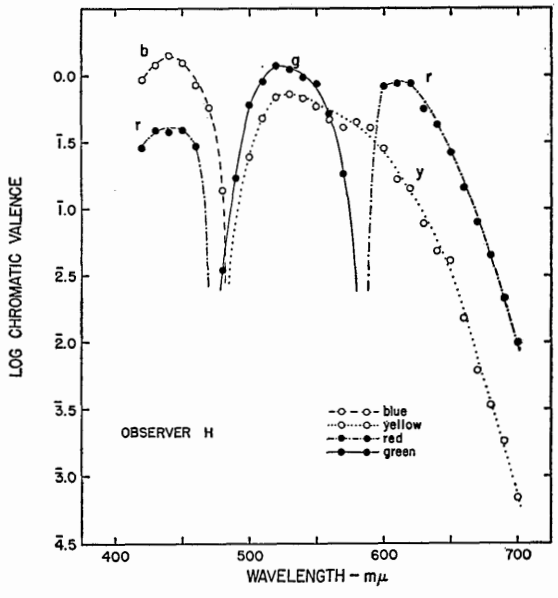

FIG. 2. Log chromatic response functions. Observer *H*.

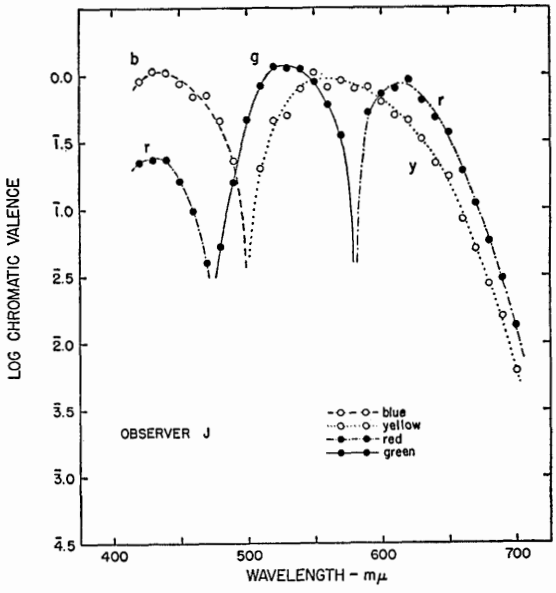

FIG. 3. Log chromatic response functions. Observer *J*.

Two determinations of the energy of the cancellation stimulus were made at each wavelength of test stimulus used in a single experimental session. The average of the two settings constitutes a single measurement. Only a single spectral range containing one of the four primary hues was explored in a single experimental session. For each observer, measurements of each of the four response functions were obtained in ten repeated sessions. Thus, forty separate sessions constituted a complete experiment for one observer. The order in which the functions were obtained was random.

All observations were monocular. Each observer used the right eye.

RESULTS

Figures 2 and 3 show graphically the chromatic response or chromatic valence functions for observers *H* and *J*.[28] These functions were measured for an equal

[27] E. Hering, *Grundzüge der Lehre vom Lichtsinn* (Verlag Julius Springer, Berlin, 1920), p. 288 ff.

[28] Complete numerical data with a sigma values are available from the authors upon request.

brightness spectrum at 10 mL and for a neutral white adaptation of 10 mL, and the results were then converted to an equal energy spectrum. Log chromatic valence is plotted as the ordinate and wavelength in millimicrons is the abscissa. The open circles in each case represent the blue and yellow responses, the solid circles, red and green responses. The dashed line (— — —) represents blue response, the dotted line (· · ·) yellow, the red is represented by a dotted and dashed (—·—) line, and the green by a solid line. The curves have been fitted visually. The lower case letters, *b*, *g*, *y*, and *r*, just above each curve, identify the four chromatic functions. The red function has two branches, a short- and a long-wave branch.

The form of each chromatic curve is measured directly by the relative log energy of the opponent equilibration stimulus at each wavelength. In the case of the blue function, for example, the values are determined by the measured energy of the opponent yellow stimulus of fixed wavelength; in the case of the red curve, the values are measured by the energy of the opponent green stimulus, etc. The levels of the four curves relative to one another require, however, further adjustments. On the assumption that equal amounts of opponent hue responses (*y* and *b* or *r* and *g*) are present when the opponent hues are exactly canceled, the measured relative energy ratios in such a mixture of opponent stimulus pairs were used to adjust the paired chromatic valence curves.

This operation relates the blue curve to the yellow, and the red to the green. It does not, of course, relate the yellow-blue pair to the red-green pair. It is, moreover, a major thesis of our theoretical development that the latter relation is not fixed but rather that it is a specific, lawful function of luminance. The intersection loci of the two pairs can, however, be fixed empirically for a given luminance level by determining the wavelengths at which equal proportions of disparate hues are seen. Since equal ordinates are assumed to represent equal response levels, the intersection loci are isolated by determining experimentally those spectral loci where the observer reports the sensation as equal in redness and yellowness, equal in greenness and yellowness, etc. Brightness must, of course, be controlled in such an experimental determination. Our procedure was to present successive spectral stimuli separated by 10-mμ intervals all at the same luminance level, namely 10 mL. The observer simply specified the seen hue in terms of the four hue variables. Although the method does not lend itself to precise measurements, the transition points between, say, reddish-yellow and yellowish-red or between greenish-yellow and yellowish-green, are readily determinable. The *y-b* and *r-g* pairs of curves represented in Figs. 2 and 3 have been locked relative to each other for the 10-mL luminance level on the basis of these supplementary data. To a first approximation the relative ordinate values are correct both within pairs, and for the stated conditions, between pairs of response functions.

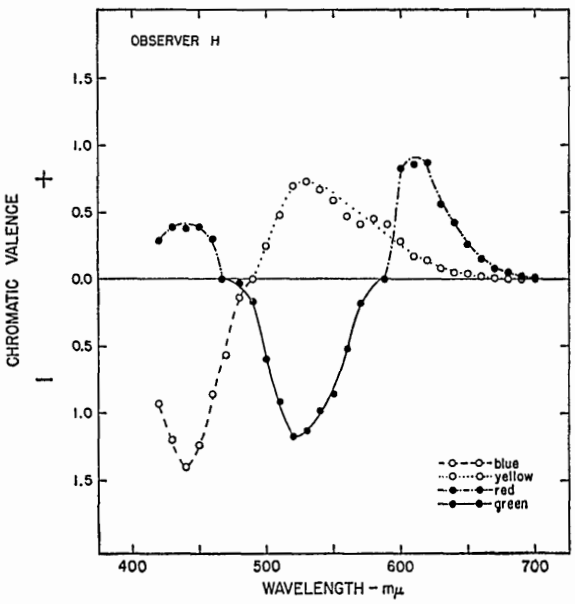

FIG. 4. Chromatic response functions. Observer *H*.

To the best of our knowledge these curves are the first such measures of chromatic response functions. Strictly hypothetical sets of so-called valence curves have appeared from time to time in the literature,[29] and the single set of functions measured by Brückner at Hering's instigation in 1903 employed only arbitrary stimulus specifications (slit widths).[21]

Generally speaking the functions for the two observers in Figs. 2 and 3 are similar. Both the forms and relative magnitudes of comparable functions tend to be alike, and the maxima and the minima appear at comparable spectral loci. Clearly, similar properties of the visual system are being measured.

A closer examination of the data, however, reveals some differences between the two observers. Although both the *b* and *g* response maxima are greater than the *y* and *r* maxima for both observers, the maximum response occurs in the blue for observer *H* while for observer *J* it occurs in the green. For both observers, the peak loci of the curves are at 440 mμ and 620 mμ for red, 430–440 mμ for blue, and 520 mμ for green. The yellow response, however, is maximal at 530 mμ for observer *H*, whereas it peaks at 560 mμ for observer *J*.

The results plotted in Figs. 2 and 3 have been converted to arithmetic units and the opponent members of each chromatic pair have been given arbitrary positive and negative specifications to correspond with these opponent characteristics. In this way, the two separate functions for the two members of each chromatic response pair are reduced to a single function with positive and negative branches. The arithmetic functions are presented in Figs. 4 and 5. The symbols and lines all

[29] A. v. Tschermak-Seysenegg, Naturwiss. 18, 589 (1930).

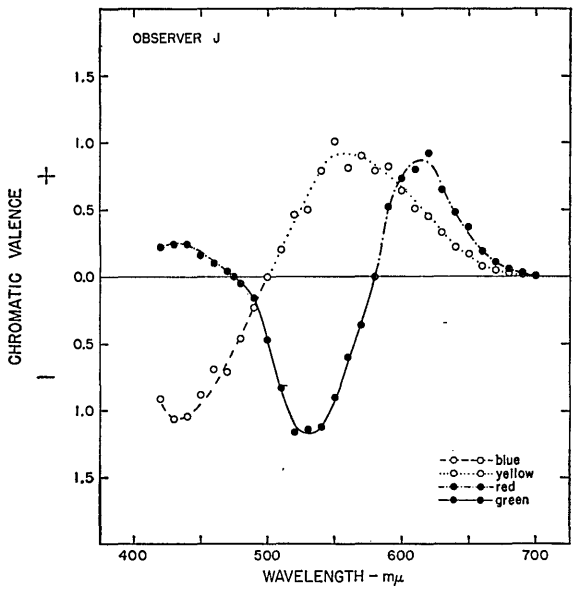

FIG. 5. Chromatic response functions. Observer *J*.

have the same significance as in Figs. 2 and 3. The intersection loci of the *r-g* and *y-b* curves with the zero response line correspond with the wavelengths at which pure or unique hues are seen. For observer *H* the pure blue is located at 467 mμ (red and green are both equal to zero), the pure green is at 490 mμ (blue and yellow are both equal to zero) and the pure yellow occurs at 588 mμ (zero value for both red and green). For observer *J* the pure blue, green, and yellow loci are at 475 mμ, 500 mμ, and 580 mμ, respectively. A point of special interest is the fact that the magnitude of the response of the chromatic process correlated with a given hue is in no instance maximal at the wavelength locus where that hue is pure or unitary and where all the remaining chromatic responses are equal to zero. This finding confirms the prediction made by Hering in one of his early theoretical papers.[30]

Since perceived hue is the criterion used to measure the chromatic response functions, for the specified conditions these functions (unlike the usual sets of trichromatic response curves that represent color mixture data) correlate directly with perceived hue. (See Wright[31] and Thomson[32] on the failure of trichromatic mixture curves to describe adequately the quality of the sensations.) Consider, e.g., Fig. 5. Starting at the low wavelength and moving to higher wavelengths the response functions indicate the hues to be reddish-blue (violet), pure blue (475 mμ), greenish-blue, bluish-green, pure green (500 mμ), yellowish-green, greenish-yellow, yellow (580 mμ), reddish-yellow, and finally yellowish-red. Yellow is seen to be present throughout the long-wave region up to 700 mμ, thus confirming the reports of Helmholtz and others that even the extreme spectral reds are slightly yellowish in hue.[33] In a later paper of this series we shall discuss in greater detail the correlation between perceived color and the measured chromatic responses.

Apart from the descriptive advantage of the chromatic response functions presented in Figs. 4 and 5, the general usefulness of such functions can best be judged in terms of their subsumptive and predictive powers.

Consider, for example, the saturation-wavelength function discussed in our introductory remarks. Figure 6 contains saturation discrimination functions obtained in four separate experiments for eight observers.[4–7] These functions were obtained by adding, in all instances, monchromatic light to white. Ordinate values represent the log of the reciprocal of colorimetric purity, abscissa is wavelength. The different sets of functions have been arbitrarily displaced along the ordinate for clarity of presentation. Reading from top to bottom in Fig. 6, the experimental functions are those obtained by Wright and Pitt; Nelson; Priest and Brickwedde; and Martin, Warburton, and Morgan. The observer's initials are indicated to the right of each curve. The data obtained by Grether[8] and Chapanis[9] are very similar to those reproduced in the figure.

As already indicated, all the experimental results are similar: the spectral saturation functions show maxima in the short- and long-wave regions and a sharp minimum in the region of 570 mμ. For the more extended

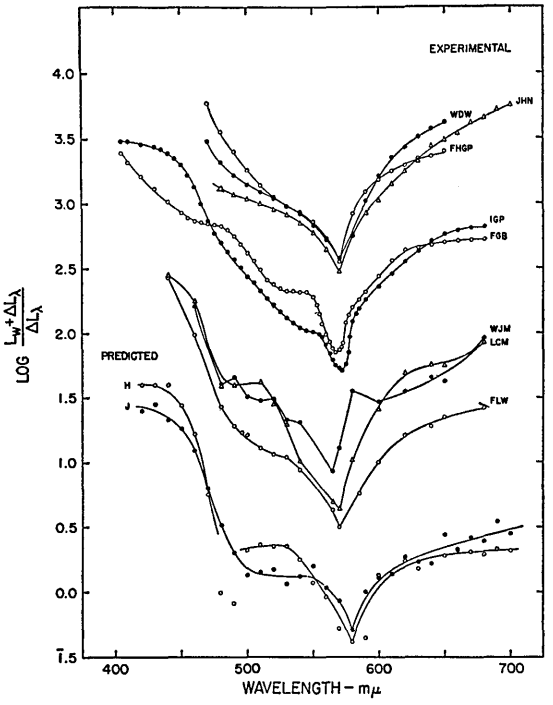

FIG. 6. Spectral saturation functions (see text).

[30] E. Hering, Lotos, Jahrb. Naturwiss. N.F. **1**, 81 (1880).
[31] Reference 15, pp. 165–166.
[32] L. C. Thomson, Ophthal. Lit. **6**, 14 (1952).
[33] H. v. Helmholtz, *Handbuch der Physiologischen Optik* (L. Voss, Hamburg and Leipzig, 1896), second edition, p. 456.

functions, the short-wave maximum is higher than the maximal value in the long-wave region.

The experimental method used to measure saturation discrimination assumes that the threshold increment in a given spectral light will vary inversely with the ratio of chromatic to achromatic components in the sensory response to that light. Since our measured chromatic response functions are assumed to provide an independent measure of chromatic excitation, and since measured luminosity functions obtained earlier for the same two observers[23] provide an independent measure of the achromatic response, we have the necessary data for predicting the saturation function for our observers. Logarithmic functions based on the ratio of chromatic/achromatic response at each wavelength[34] are presented for observers *H* and *J* in the lowest two curves in Fig. 6. Two points in the function for observer *H* remain unconnected in this visual fit. Direct measures of the saturation function for this observer will alone ultimately determine the validity of the secondary inflection point indicated by these two values. The close approximation of the predicted to the experimental functions is, nevertheless, obvious.

There remains no doubt that chromatic and achromatic responses can be measured independently, and that the measurements can be used to account for variations in saturation discrimination with changes in wavelength. It therefore seems fruitful to pursue the quantification and generalization of an opponent-colors theory, which assumes separate chromatic and achromatic responses, in order to provide an integrated account for some of the more important facts of color perception and discrimination. The theoretical model will be developed and examined in the second paper of this series.

[34] Reference 30, p. 77.

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Determination of Hg^{202} and Other Mercury Isotopes in Samples of Mercury Vapor by Mercury Resonance Radiation Absorbiometry*

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(Received September 3, 1954)

A method is described for the determination of the percentage of Hg-202 present in small samples of mercury. An electrodeless discharge tube containing Hg-202 is used as a source of the Hg-202 hyperfine component of the 2537-A resonance line of mercury. By determining the absorbance of this component by mercury vapor, the quantity of the isotope present in the vapor can be determined. The analyses of samples of mercury containing various percentages of Hg-202 agree with values obtained by mass spectrometric methods to within two percent. The method has the advantage over the mass spectrometer of requiring very much smaller samples. Furthermore, contamination from "hangup" of previous samples is avoided. Through the use of other isotopic sources it is shown that the method could be extended to the analysis of the remaining isotopes of mercury.

THE isotopic analysis of samples of mercury by means of the mass spectrometer is complicated by the tendency of the vapor to "hang up" in the inlet system of the instrument. Under such conditions, succeeding samples will show contamination due to adsorbed mercury from previous analyses. The situation becomes particularly serious when the quantity of sample available is small.

During the course of an investigation in this Laboratory of the mercury-6($^3P_1$)-photosensitized decomposition of isopropyl chloride, in which an Hg-202 electrodeless discharge tube was used as the source of resonance radiation, it became necessary to determine the atom-percent Hg-202 in the mercury present in the calomel product of the reaction. A method has been developed for this purpose, involving the absorption of resonance radiation, by means of which analyses can be made on microgram quantities of mercury rapidly and with good precision. Furthermore, it will be shown that the method can be easily extended to the determination of other mercury isotopes.

The hyperfine structure of the λ2537-A resonance line of mercury has been shown to consist of five resolvable hyperfine lines. Some of these lines arise from a single even-mass isotope, while others are formed from the superimposition of components from several isotopes. The pertinent data[1] are given in Table I.

It will be noted from the table that the *X*- and *Y*-components arise exclusively from Hg-200 and Hg-202, respectively. Hence, if a beam of either of these components is allowed to traverse mercury vapor, the fractional absorption will be a function of the number of atoms of the corresponding isotope present in the vapor.

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[1] J. L. Cojan and R. Lennieur, Compt. rend. **235**, 1634 (1952); K. Burns and K. B. Adams, J. Opt. Soc. Am. **42**, 56 and 716 (1952).