

# P-1—The Laser Absolute Wavelength Standard Problem

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**Abstract**—Stabilized lasers usually exhibit systematic frequency shifts larger than their resettability; this phenomenon is well illustrated by the 6328-Å helium–neon laser. We describe a Lamb-dip stabilized laser that operates at 1.15259  $\mu$  in pure low-pressure (0.12-torr) neon. Optical heterodyne experiments indicate an accuracy exceeding 1 part in  $10^9$ ; short and medium term precisions of  $1:10^{10}$  are easily achieved. We also report the successful operation of a wavelength reference based on the saturation of sharp molecular absorption. In the first experiments the  $P(7)$  line of the  $\nu_3$  band of methane is saturated inside the cavity of a 3.39- $\mu$  helium–neon laser. The saturation maximum at molecular line center produces an “emission” feature whose linewidth is less than 5 parts in  $10^6$ . The pressure-induced offset is expected to be less than 1 part in  $10^{10}$ . Size scaling is expected to improve these first results by at least 1 decade.

SINCE the days when negative temperature atomic transitions were first available in the visible and near-infrared, much thought has been invested in the potential application of oscillator devices as wavelength standards. The Shawlow–Townes spectral width relationship [1] predicts intrinsic oscillating bandwidths of the order of a hertz or less for essentially all gas lasers. The work of Jaseja *et al.* [2] showed that with (much) care one could reach oscillating bandwidths of a few tens of hertz, limited primarily by mechanical and thermal disturbances of the environment. This spectral width corresponds to an uncertainty in the instantaneous frequency of less than 1 part in  $10^{13}$ . By contrast, the resettability of a given frequency was reported [3] as about  $1:10^9$  some four decades before one begins to confront problems of a fundamental nature.

Since there is not much published work dealing with absolute resettability, we illustrate the problem of constructing an absolute laser wavelength standard by considering briefly what can be done in the visible.

We recognize at the outset that ordinarily the cavity  $Q$  will exceed operating atomic  $Q$  by a factor of at least 50 and that consequently a servomechanism will be required, which continuously resets the cavity resonance to some fiducial mark in the atomic profile. For the argument, we will assume a servo that uses the absorption (or dispersion) part of the *natural width*, rather than the Doppler width. From a 20-cm 1.35-torr single-frequency 6328-Å helium–neon laser, we have obtained 250  $\mu$ W with a symmetric Lamb dip of 30 percent depth and 110-MHz FWHM [4]. This linewidth, which is a fractional linewidth of  $\pm 1.1$  parts in  $10^7$ , can easily be split into, say, 100 or 200 parts,

giving an uncertainty of less than 1 in  $10^9$ . However, the pressure induced offset [5] is about + 25 MHz, or + 5 parts in  $10^8$ . Thus the laser gas pressure and composition would have to be known to better than 5 percent to make the accuracy comparable to the reasonably good resettability. As an illustration of this large gap between *precision* and *accuracy* we mention the recent intercomparison [6] by three national standards laboratories of the wavelength of a single commercial 6328-Å self-stabilized oscillator. Since the intercomparisons were accomplished in a short time, compared with the operating lifetime of the device, we are not surprised that the results agreed within  $\pm 5$  parts in  $10^9$ . In fact, the precision of this intercomparison is probably limited as much by the properties of the krypton wavelength standard as by the laser itself. However, the critical feature is that another laser of similar manufacture exhibited a center wavelength offset from the first by more than 1 in  $10^7$  due to different operating parameters, principally pressure. We conclude that the combination of strong pressure shifts, moderately high operating pressures, and a multicomponent gas medium effectively rules out the 6328-Å self-stabilized oscillator as a primary standard.

There have been a number of ingenious proposals put forward to improve the sensitivity with which to detect deviations of oscillation frequency from the center of the laser natural line. These concepts include among many others: 1) scanning-type Lamb-dip amplitude stabilizers [7]; 2) dispersion stabilizers based on frequency shifts induced by gain modulation [8] or by loss modulation [9]; 3) the bistable polarization property of certain weak-field Zeeman lasers near line center [10]; 4) magnetically-induced circular dichroism [11]; and 5) reduction in the  $c/2L$  photobeat when the FM laser is tuned through line center [12]. Some of these techniques may well prove important in the achievement of good short-term stability in unfavorable environments. However, at present there is little doubt that the basic problems in achieving a useable laser wavelength standard relate to *systematic* effects and not to signal-to-noise ratios. Thus, in the work to be described, scanning-type amplitude stabilizers are employed. This method is less complex and, more importantly, may be definitively tested for systematic instrumental bias.

Of the many properties desired in a primary wavelength standard, such as convenient wavelength, adequate power output, simplicity and economy, long life, and excellent short-term stability, there is, by definition, nothing so important as good long-term stability. In view of the good signal-to-noise ratio attainable with lasers, we expect to be able to design a servomechanism that makes the oscillation frequency approach the natural frequency very closely

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indeed. However, the molecules or atoms, whose transitions we use as the reference element, experience electric fields (Stark shifts), magnetic fields (Zeeman shifts), thermal and macroscopic velocity fields (Doppler shifts), and pressure-induced line broadening and shifts. Any meaningful investigation of potential absolute standards must consider these phenomena, as well as other systematic instrumental effects. We can well hope to find a transition that is only weakly affected by these perturbations, eliminate as many perturbations as possible, and then provide a standard reproducible operating point for the others. Finally, by taking advantage of the excellent medium-term stability, we should be able to "extrapolate off" the most serious remaining shifts.

It has long been realized that a single-gas component system must be used in any potential laser wavelength standard. However, this prerequisite for accuracy may well preclude the realization of any gain at the optical frequency. Consequently single-gas absorption cells have been used both inside and outside laser cavities in conjunction with a regular amplifier discharge tube [13]. We may treat the work of Lee and Skolnik [14] as an example. These workers put a pure neon absorber, as well as a helium-neon gain cell, inside the laser cavity, thus increasing their sensitivity to small absorptions. Their 6328-Å oscillator made sufficient power to partially saturate the slightly broadened natural line ( $\sim 15$ -MHz width) of the neon absorber. At the absorber line center, where the two holes burned in the velocity distribution maximally overlap, the saturation of the absorption is more complete, and thus an increased laser output results. This trick largely overcomes the deficiencies of the 6328-Å He-Ne oscillator and an eminently useful working standard should result. Ultimately, due to Lamb-dip effects in the gain cell coupled with the large ( $\sim 60$ -MHz) pressure-induced offset of the amplifier cell, it may be expected to be difficult to obtain the symmetric and bias-free discriminant necessary to explore the range substantially below  $1:10^9$ .

As a second example, we report on work being carried out at the Joint Institute for Laboratory Astrophysics in collaboration with Dr. H. S. Boyne of the National Bureau of Standards. We study the  $1.15259$  ( $2S_2 \rightarrow 2P_1$ ) neon line, which oscillates at low pressures in pure neon as first reported by Bennett *et al.* [15]. In 20-cm discharge tubes (2-mm bore), just a few watts of VHF power provide gains exceeding 1 percent, giving an output power of 5 to 10  $\mu$ W through a 1.5 percent transmission mirror. At 120 to 150 mtorr of neon, the Lamb dip is about 45 MHz wide and deeper than 30 percent. Fig. 1 shows this output at several excitation levels, the lowest one corresponding to about 3 cm of discharge. The output mirror in this case was 99.5 percent reflecting, which results in a rather substantial loss of attainable single-frequency power compared with optimal coupling.

For an amplitude-measuring type of stabilizer, the symmetry of the Lamb dip is of critical importance. We believe the folded confocal cavity should be used with the smallest bore tube possible to provide very large loss for

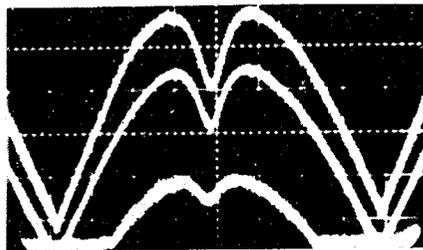


Fig. 1. Power output of  $\lambda = 1.15 \mu$  single-frequency pure neon laser versus cavity tuning. The very slight asymmetry visible was affected by cavity alignment and is thought to arise from the admixture of off-axis modes. Neon  $^{20}$ :  $p=120 \mu$ ;  $c/2L=456$  MHz;  $P=1 \mu$ W/div.

off-axis modes. The low  $Q$  and intrinsic frequency degeneracy of these cavity modes allows one to obtain a very symmetric and bias-free discriminant. The symmetry of the Lamb-dip region may be studied in detail by the technique of "frequency offset locking." With this method the stability of an optimized laser, stabilized at line center (with, say, a scanning-type Lamb-dip stabilizer), may be transferred to another laser operating under more general conditions. The second laser cavity is servo-driven to produce a photobeat frequency equal to a laboratory RF signal generator. Thus one is able to eliminate the influence of cavity vibrations and at the same time provide an extremely high-resolution adjustable frequency offset of the second laser from line center (as defined by the first locking servo). In some preliminary experiments, using an analog form of frequency offset locking, we have studied the slope of the Lamb dip of the offset laser near zero beat (see Fig. 2). An extrapolated beat frequency of  $(210 \pm 15)$  kHz at the zero-slope point was obtained, corresponding to an offset of  $(8.1 \pm 0.6) \times 10^{-10}$ . Increasing the pressure in the offset laser by 100 mtorr (to 290-mtorr total) resulted in an offset of  $(8.1 \pm 0.7) \times 10^{-10}$ . We conclude that the laser frequency is remarkably insensitive to pressure. (Also the Lamb dip is observed not to broaden measurably with pressure, even up to 0.4 torr.) The total offset from the free atom frequency is almost surely less than  $1 \times 10^{-9}$ . Using the ambipolar diffusion model of Mayer [16] and the electron temperature and density measurements of Gordon and Labuda [17], we can estimate the total Stark shift of this line to be of the order of  $+10$  kHz ( $+4 \times 10^{-11}$ ). Present work is aimed at understanding the origin of the experimental 200-kHz offset. Since one of the lasers used had too large a bore, long radius mirrors were used. It is surmised that the off-axis modes that were observed at high excitations caused the  $Q$  of the cavity to be dependent upon its tuning. The experiments will be repeated with more nearly optimized lasers.

Given a well designed laser with reasonable short-term stability, signal-to-noise ratios for the discriminant are not yet of primary concern. However, it still would be interesting to see if the apparently large population density of the  $2S_2$  level in pure low-pressure neon would allow oscillation on the  $2S_2 \rightarrow 2P_1$   $1.52349\text{-}\mu$  line. This transition is of the  $J = 1 \rightarrow J = 0$  type, and consequently should give the extremely sharp discriminant obtainable from the bistable

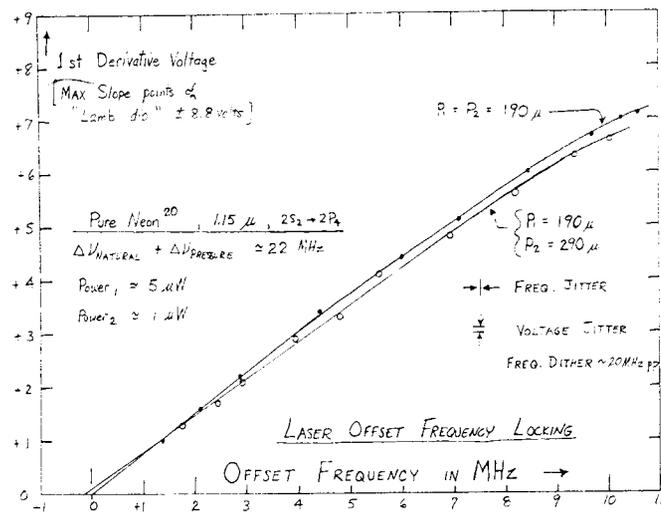


Fig. 2. Derivative of output power versus frequency, measured from the Lamb-dip minimum. This laser was servo controlled by photobeating with a Lamb-dip stabilized local oscillator. The frequency jitter of  $\pm 10$  MHz was thereby transferred to the first laser. The variation of power was then synchronously rectified to measure one side of the Lamb-dip derivative function. The noise of  $\sim 10$  kHz rms in the offset frequency contributes essentially nothing to the observed 0.1-volt slope jitter.

polarization characteristics of a Zeeman laser as discussed by Fork *et al.* [18] and Polanyi *et al.* [19].

For reference, in Table I, we summarize the properties of the 1.15- $\mu$  pure neon Lamb-dip stabilized wavelength reference. Although its performance is not as good as that of the saturated absorber system to be described presently, its simplicity, low power requirements, accuracy, and stability commend it highly as a working standard.

In all likelihood, the "ultimate" wavelength standard will be based on sharp-line absorption of laser light by suitable molecules [20]–[24]. In the provocative work of Ezekial and Weiss [24], a molecular beam of iodine was observed to fluoresce when illuminated with light near 5145 Å. Electric quadrupole interactions appear to give resolvable structure to the fluorescence excitation profile. Lifetime measurements indicated a potential interaction time approaching 3  $\mu$ s, corresponding to less than a 100-kHz linewidth. Unfortunately, the Ar<sup>+</sup> laser requires high-power inputs ( $\sim$  kilowatts) and it may therefore be expected that good short-term stability will be rather difficult to achieve. To eliminate first order Doppler broadening, the molecular beam will need to be collimated within about 20 arc seconds and perpendicular to the double-passed laser beam to the same accuracy. These are basically experimental considerations and it is by far too early to predict the outcome.

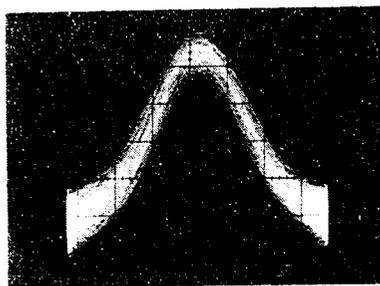
Several other examples of laser-molecular absorption coincidences are known, including various alkali molecules with argon<sup>+</sup> and 6328-Å neon laser lines [21]–[25], NO<sub>2</sub> with several argon<sup>+</sup> lines [26], etc. The classic case—and probably the most exciting at the moment for standards purposes—is the 3.3913- $\mu$  He-Ne laser pumping the P(7) line of the  $\nu_3$  band of CH<sub>4</sub>. Earlier absorption work on this case was reported by Gerritsen [27]. Use of the narrow

TABLE I  
A USABLE Interim LASER WAVELENGTH REFERENCE

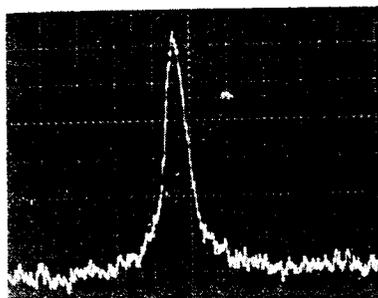
Pure neon <sup>20</sup>	11 525.9 Å	(2S <sub>2</sub> → 2P <sub>2</sub> )
Electron impact pumping: $pd \approx 0.3$ torr mm.		
Operating pressure range: 80 to 400 mtorr.		
Optimum pressure: 120 to 150 mtorr.		
Power input to RF oscillator (170 MHz): 9 watts dc.		
Estimated input to discharge $\sim 4$ watts.		
Gain in 20-cm tube (2-mm bore) $> 3$ percent.		
Power output through 2 percent transmitting mirror $> 5 \mu$ W.		
Lamb-dip width $\sim 22$ MHz FWHM.		
Natural width $\approx 15$ MHz FWHM.		
Lamb-dip depth $> 30$ percent.		
Stark shift at operating point a) $\sim +6$ kHz ( $+3 \cdot 10^{11}$ ).		
Pressure shift at operating point b) $< 200$ kHz ( $< 8 \cdot 10^{10}$ ).		
Estimated accuracy (total offset from free atom frequency) $< 1 \cdot 10^9$ .		
Demonstrated precision (medium term stability) $< 1 \cdot 10^{10}$ .		
a) Calculated value, averaged over mode size.		
b) Quoted as entire offset between 2 independent devices, variation with pressure appears to be substantially less than this value.		

Doppler absorption profile of saturated CH<sub>4</sub> vapor held at the nitrogen triple point (63.2°K) has been suggested as a wavelength reference by Shimoda [28]. In collaboration with Dr. R. L. Barger, we have been applying the technique of saturated absorption to the methane  $-3.39\text{-}\mu$  case. As pointed out by Lee and Skolnick [14], an emission feature of width related to the natural (rather than the Doppler) width occurs when the laser frequency is in resonance with atoms having only transverse velocity components. For the CH<sub>4</sub> case, at 10-mtorr gas pressure we obtain in preliminary experiments an emission feature about 2 percent of the basic laser intensity (see Fig. 3). This symmetric peak, approximately Lorentzian in form, has a spectral width of less than 1.5 MHz! This width is dominated almost entirely by the finite molecular time of flight through the aperture of the saturating laser beam. When the molecular velocity is reduced by cooling the gas to 77°K, the linewidth is reduced to 660 kHz FWHM. With a larger aperture we expect to more nearly approach the 75-kHz pressure-induced linewidth [27] characteristic of the 10-mtorr operating pressure. The peak can be easily seen down to about 1 mtorr; at this pressure the pressure-induced linewidth is 7.5 kHz and the offset is expected to be less than  $1 \cdot 10^{11}$ . From the absorption coefficient and linewidth, the radiative linewidth is calculated to be only about 100 Hz. With an ordinary InAs photodiode we obtain a signal-to-noise ratio of about 6000:1 with a 1-Hz bandwidth. Virtually all the observed noise is due to the poorly matched amplifier input stage.

It is entertaining to speculate on the potential of this saturated molecular absorption approach to the wavelength standard problem. Two versions of the 10- $\mu$ W pure neon laser previously described stabilize themselves independently at the line center to within  $\pm 1/500$  of the full width. Taking this fraction as a demonstrated and realistic guide (likely pessimistic in view of the 100-fold-greater power output of the 3.39- $\mu$  device), we anticipate a precision of about 3 parts in  $10^{12}$  for model II, presently being constructed. The accuracy will likely be affected by factors



(a)



(b)

Fig. 3. Saturated molecular absorption signal from methane cell inside (a) 3.39- $\mu$  He-Ne laser cavity. The peak, due to saturation of the opacity of molecules traveling transverse to the laser beam, is about 2 percent of the 1-mW laser output. Its width of 1.5 MHz is due to the short time of flight across the effective mode-spot size. Both refrigeration of the gas to 77°K and increase of mode diameter give their anticipated linewidth narrowing. Pressure-induced width is about 75 kHz. Laser cavity locked to the saturated absorption resonance. (b) Same conditions as above, except with wider sweep to show wings and noise level. Display bandwidth is 4.5 kHz. Cavity is unlocked.

not yet fully appreciated, but as pointed out by Shimoda [28], the relevant line of  $\text{CH}_4$  is free from measurable Stark shift. Zeeman splitting in the earth's field should be about 500 Hz. Nuclear spin-rotation interaction is expected to be less than 1 kHz. If this structure can be properly taken into account, it may be possible to define a meter that would rival the second in terms of its reproducibility.

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#### REFERENCES

- [1] A. L. Schawlow and C. H. Townes, "Infrared and optical masers," *Phys. Rev.*, vol. 112, p. 1940, December 1958.
- [2] T. S. Jaseja, A. Javan, and C. H. Townes, "Frequency stability of He-Ne masers and measurements of length," *Phys. Rev. Letters*, vol. 10, p. 165, 1963.
- [3] T. S. Jaseja, A. Javan, J. Murray, and C. H. Townes, "Stability and resettability of He-Ne masers," *Bull. Am. Phys. Soc.*, vol. 7, p. 553, November 1962.

- [4] This result agrees well with predictions based on the work of P. W. Smith, "Linewidth and saturation parameters for the 6328 Å transition in a He-Ne laser," *J. Appl. Phys.*, vol. 37, p. 2089, April 1966.
- [5] A. L. Bloom and D. L. Wright, "Pressure shifts in a stabilized single wavelength helium-neon laser," *Appl. Opt.*, vol. 5, p. 1528, October 1966. Also P. Sosnowski and W. B. Johnson, "Frequency shifts of the Lamb dip minimum in the helium-neon laser," presented at the 1968 Internat'l Quantum Electronics Conf., Miami, Fla., paper 16P-10. Abstract in *IEEE J. Quantum Electronics*, vol. QE-4, pp. 370-371, May 1968. Also A. D. White, "Pressure and current dependent shifts in the center frequency of the Doppler-broadened ( $2P_4-3S_2$ ) 6328 Å Ne transition," *Appl. Phys. Letters*, vol. 10, p. 24, 1967.
- [6] K. D. Mielenz, K. F. Nefflen, W. R. C. Rowley, D. C. Wilson, and E. Engelhard, "Reproducibility of helium-neon laser wavelengths at 633 nm," *Appl. Opt.* vol. 7, p. 289, February 1968.
- [7] Various authors following related work by W. R. C. Rowley and D. C. Wilson, "Wavelength stabilization of an optical maser," *Nature*, vol. 200, p. 745, 1963.
- [8] W. R. Bennett, S. F. Jacobs, J. T. La Tourrette, and P. Rabinowitz, "Dispersion characteristics and frequency stabilization of a gas laser," *Appl. Phys. Letters*, vol. 5, p. 56, August 1964.
- [9] G. Luther and Z. Bay, NBS, Gaithersburg, Md., private communication.
- [10] See, for example, [18] and [19].
- [11] A. D. White, "Frequency stabilization of gas lasers," *IEEE J. Quantum Electronics*, vol. QE-1, pp. 349-357, November 1965.
- [12] S. E. Harris, "Proposed frequency stabilization of the FM laser," *Appl. Phys. Letters*, vol. 7, p. 185, October 1965.
- [13] See, for example, [11] and [14].
- [14] P. H. Lee and M. L. Skolnick, "Saturated neon absorption inside 6328 Å laser," *Appl. Phys. Letters*, vol. 10, p. 303, 1967.
- [15] W. R. Bennett, Jr., and J. W. Knutson, Jr., "Simultaneous laser action on the neon doublet at 1.1523  $\mu$ ," *Proc. IEEE (Correspondence)*, vol. 52, pp. 861-862, July 1964.
- [16] F. J. Mayer, "Stark effect produced frequency shifts in the helium-neon laser," *IEEE J. Quantum Electronics (Correspondence)*, vol. QE-3, pp. 690-691, December 1967. Unfortunately this author did not evaluate his model in complete detail.
- [17] E. F. Labuda and E. I. Gordon, "Microwave determination of average electron energy and density in He-Ne discharges," *J. Appl. Phys.*, vol. 35, p. 1967, May 1964.
- [18] R. L. Fork, W. J. Tomlinson, and L. J. Meilos, "Hysteresis in a helium-neon laser," *Appl. Phys. Letters*, vol. 8, p. 162, April 1966.
- [19] T. G. Polanyi, M. L. Skolnick, and I. Tobias, "Frequency stabilization of a gas laser," *IEEE J. Quantum Electronics (Correspondence)*, vol. QE-2, pp. 178-179, July 1966.
- [20] K. M. Baird, National Research Council, Ottawa, Canada, private communication.
- [21] R. N. Zare, Joint Institute for Laboratory Astrophysics, Boulder, Col., private communication.
- [22] G. Gould, P. Rabinowitz, and J. T. La Tourette, Polytechnic Institute of Brooklyn, Brooklyn, N. Y., private communication.
- [23] V. P. Chebotayev, S. N. Bagaev, Yu. D. Koloranikov, and V. N. Lisitsyn, "Frequency stabilization of gas lasers," presented at the 1968 Internat'l Quantum Electronics Conf., Miami, Fla., paper 16P-4. Abstract in *IEEE J. Quantum Electronics*, vol. QE-4, p. 368, May 1968.
- [24] S. Ezekial and R. Weiss, "Laser-induced fluorescence in a molecular beam of iodine," *Phys. Rev. Letters*, vol. 20, p. 91, 1968.
- [25] M. McClintock and L. C. Balling, "Laser excitation of atomic and molecular fluorescence spectra in cesium and rubidium," *Bull. Am. Phys. Soc.*, vol. 13, p. 55, January 1968.
- [26] H. P. Broida, post-deadline paper, presented at the 1968 Internat'l Quantum Electronics Conf., Miami, Fla.
- [27] H. P. Gerritsen, "Tuned-laser spectroscopy of organic vapors," in *Third International Conference on Quantum Electronics*, P. Grivet and N. Bloembergen, Eds. New York: Columbia University Press, 1964, p. 581.
- [28] K. Shimoda, "Absolute frequency stabilization of the 3.39  $\mu$ m laser on a  $\text{CH}_4$  line," presented at the 1968 Conf. on Precision Electromagnetic Measurements, Boulder, Col. I am particularly grateful to Prof. Shimoda for making a preprint of his work available before publication.