double-quantum transition ($2S_{1/2}$–$3S_{1/2}$). The shift of this transition can be investigated by Doppler-free rf-optical double-quantum laser spectroscopy[7] which uses the saturated polarization method and is described in Ref. 3.

In addition to the buffer gas effects on the $H_\alpha$ lines, a shift and broadening due to the microscopic electric fields of the charged particles is observed, both of which are proportional to the discharge current (50 to 500 mA), e.g., $\Delta\nu_m = -1.3(1)$ and $\Delta\nu_{1/2} = 18(2)$ in MHz/100 mA at $p_{He} = 0.1$ Torr (0 °C). By measuring the proportionality of current to electron or ion densities by means of Langmuir probes the shift-density relation is obtained.

Thus values for the displacement and broadening of all five H $n = 3$ fs levels due to collisions with buffer gas atoms and charged particles will be known and therefore be accessible to a comparison with theory. The $n = 2$ levels are more than an order of magnitude less sensitive to the above perturbations than the $n = 3$ states.

By means of a convolution integral of the shifts and broadening of the individual fs components it will be possible to calculate accurate values of the integral shift and broadening for the blended $H_\alpha$ line by taking into account the influence of collision-induced sensitized fluorescence. This integral shift is of course observed in astrophysics and in all laboratory experiments subjected to Doppler broadening.

Applications of the reported method to deuterium and other buffer gases than He, in particular H and $H_2$, are planned, as well as using the technique to measure charged-particle densities in higher-density plasmas.

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### S.6. High-Resolution Saturated Absorption of the $^1S_0$–$^3P_1$ Calcium Transition in an Atomic Beam,

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In our continuing studies[1,2] of the feasibility of the $^1S_0$–$^3P_1$ 6573 Å transition of calcium as a visible wavelength/frequency standard, we are now able to report linewidths as narrow as 50 kHz. The superior properties of this transition which make it a promising candidate for a $\lambda/\nu$ standard have been reported elsewhere.[3] Briefly, these include a nondegenerate ground state, no hyperfine structure, and a long lifetime of 0.39 ms (permitting an ultimate fractional linewidth of about $10^{-12}$, with corresponding reproducibility and accuracy exceeding $10^{-15}$). We expect to soon resolve the recoil splitting (23 kHz) which will remove the doublet structure limitation on accuracy.

In this experiment, light from a frequency-stabilized, tunable dye laser is sent at right angle through the calcium beam and retroreflected to produce the Doppler-free saturation peak. The light beam is expanded to give a 4 cm interaction length; however, experimental difficulties have so far limited our resolution to 50 kHz, rather than the expected 10 kHz time-of-flight linewidth. Perturbations on the line shape by extraneous magnetic fields are eliminated by using a field of 2–3 × $10^{-4}$ T (a few gauss) in the interaction region. This splits the $\Delta mj = \pm 1$ components away from the observed $\Delta m_j = 0$ component, which has no first-order Zeeman effect and whose second-order shift is only about $10^8$ Hz/T$^2$ (1 Hz/G$^2$). This signal is obtained by observing the fluorescence from the $^3P_1$ state downstream from the interaction region. Currently we use a 5-cm-diam cathode photomultiplier located approximately 20 cm downstream and in close proximity to the calcium beam. With this detection scheme, we estimate that we collect about 1% of the emitted photons; even so, useful S/N ratios are obtained. More efficient fluorescence detection systems are possible.[2]

Our present work also includes preliminary investigation of three zone saturated absorption optical Ramsey fringes[4,5] for this same calcium transition. With this Ramsey method, we have also achieved linewidths as narrow as 50 kHz. The important advantages of this technique include line narrowing without the degradation of signal-to-noise inherent in the usual saturated absorption spectroscopy. Power broadening or shift can be minimized. Also there is an important relaxation in the requirement of superior-quality large-aperture optics. The Ramsey method promises to be of great importance for the full realization of the potential of calcium as a $\lambda/\nu$ standard.

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### S.7. Linear Optical "Ramsey" Resonance by Means of a Spacially Modulated Molecular Beam,

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In high-resolution laser spectroscopy the attainable linewidth is ultimately limited by the finite interaction time of the molecule with the electromagnetic field. For a free molecule, because of its thermal velocity, this time is closely related to the physical dimensions of the exciting field. It therefore seemed attractive to adapt the method of separated fields, originally developed by Ramsey for microwave spectroscopy, to the optical region. In microwave spectroscopy, Doppler broadening is avoided by using a standing-wave field and confining the molecules to one antinode only. At shorter wavelengths this confinement becomes im-

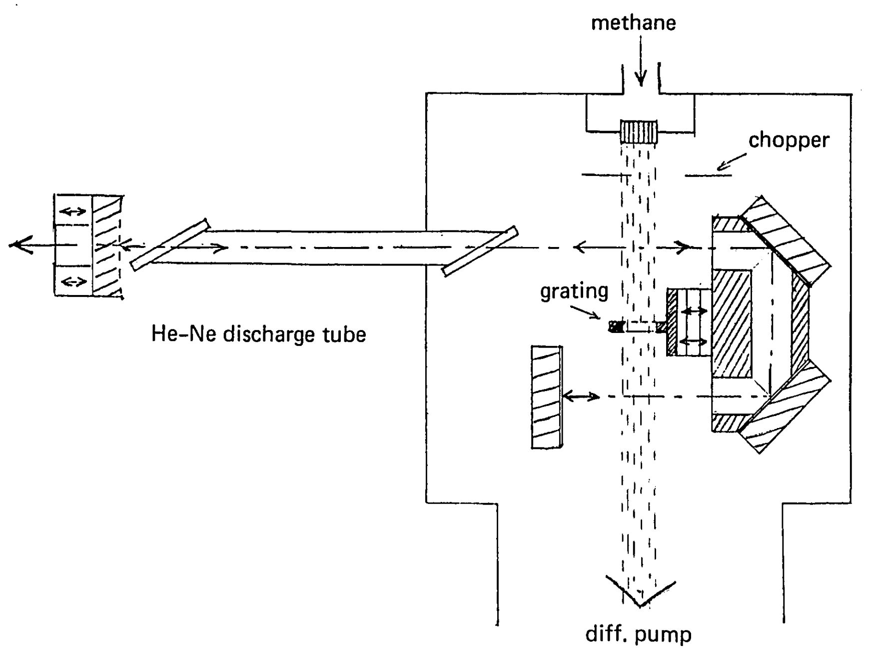


FIG. 1.