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Observation of Ramsey Fringes Using a Stimulated, Resonance Raman Transition in a Sodium Atomic Beam

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Ramsey fringes have been observed using a stimulated, resonance Raman transition at 1772 MHz excited by two dye lasers in a sodium atomic beam. The width of the central fringe is 650 Hz (half width at half maximum), corresponding to a 30-cm interaction-region separation. The fringes are free from laser jitter because the jitters in both laser beams are correlated. Applications to frequency standards as well as to high-resolution spectroscopy in the microwave to far-ir regions are discussed.

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We report the observation of Ramsey fringes using a stimulated, resonance Raman transition between two long-lived hyperfine ground sublevels, separated by 1772 MHz, in a sodium atomic beam. The observed fringes have a width of 650 Hz [half width at half maximum (HWHM)] for an interaction-region separation of 30 cm which is consistent with transit-time effects in a thermal sodium atomic beam. To our knowledge, these are the narrowest features recorded using optical lasers and have applications in high-resolution spectroscopy and in the development of new time and frequency standards in the microwave to submillimeter regions and possibly also in the far-ir region of the spectrum.[1]

Figure 1(a) shows schematically a stimulated, resonance Raman transition between two long-lived states, 1 and 3, using two laser fields, ω_1 and ω_2, resonant with the intermediate state 2. In our experiment states 1 and 3 are the $3^2S_{1/2}(F=1)$ and $3^2S_{1/2}(F=2)$ ground sublevels in atomic sodium, respectively, and state 2 is the $3^2P_{1/2}(F=2)$ level, having a 16-nsec lifetime. The Raman transition linewidth for copropagating laser fields is determined by the decay rates of states 1 and 3, with a negligible contribution from state 2, i.e., the linewidth is set by the transit time, since states 1 and 3 are long lived.[2,3] To obtain a small transit-time linewidth we use Ramsey's method of separated oscillatory fields,[4] illustrated in Fig. 1(b), with a field separation of up to 30 cm. The ability to observe high-contrast Ramsey

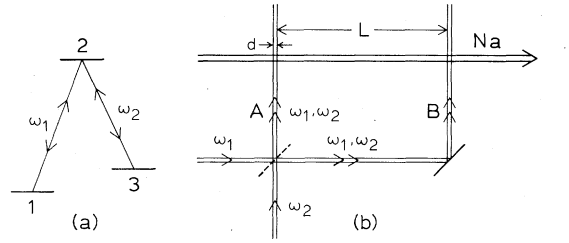

FIG. 1. (a) Stimulated, resonance Raman transition. (b) Schematic of experimental setup for obtaining Ramsey fringes.

fringes for such a separation is made possible by eliminating the effects of laser frequency jitter. This is done by generating one laser frequency directly from the other so that the difference frequency, $\omega_1 - \omega_2$, and therefore the Raman transition linewidth, is insensitive to laser jitter.[1]

Ramsey fringes induced by this Raman process are equivalent to those which would be observed in a single-step microwave transition between states 1 and 3 even though no microwaves are actually used in the interaction. This presents an attractive possibility of doing ultrahigh-resolution spectroscopy over a wide range of frequencies, from rf to far ir, using lasers. Previous observations of Ramsey fringes using lasers have been in connection with equal-frequency two-photon absorption[5,6] and also in two-level saturated absorption transitions at optical frequencies.[7-9]

Details of our experimental setup are shown in Fig. 2. The dye laser generating ω_1 is short-term stabilized by locking to a passive Fabry-Perot reference cavity and long-term stabilized by locking the reference cavity to the $1 \rightarrow 2$ transition in the atomic beam[10] as outlined in Fig. 2.

The atomic beam is collimated to about 1 mrad and the earth's magnetic field is canceled over the entire interaction length with a three-axis Helmholtz coil, not shown in Fig. 2. The laser frequency at ω_2 is derived from ω_1 by an acousto-optic frequency shifter[11] driven at approximately 1772 MHz using a microwave voltage controlled oscillator (VCO).

Both laser beams at ω_1 and ω_2 are expanded to about 3 mm $1/e^2$ diameter and collimated. They are then combined on a beam splitter to produce beams A and B as shown in Figs. 1(b) and 2. The components at ω_1 and ω_2 in both A and B beams have 16 μW of power each. Beams A and B intersect the atomic beam, Fig. 2, to give interaction regions 1 and 2, respectively, separated by a

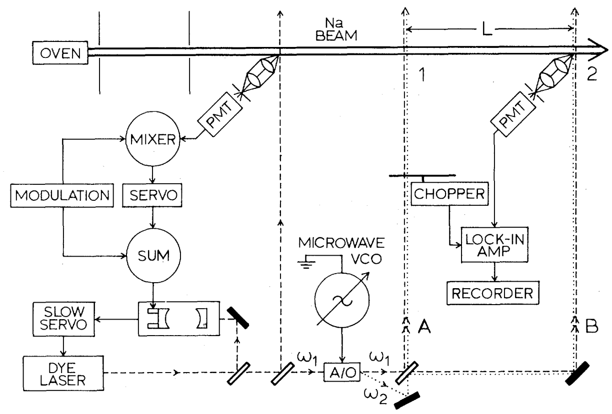

FIG. 2. Experimental setup.

distance L.

Figure 3(a) shows the stimulated, resonance Raman transition induced in region 2 with beam A blocked. Here, ω_1 is held on resonance with the $1 \rightarrow 2$ transition, and ω_2 is scanned over the $3 \rightarrow 2$ transition frequency. For convenience the polarizations of ω_1 and ω_2 are made linear but perpendicular to each other. To partially suppress the background fluorescence, ω_1 is chopped at 270 Hz (not shown in Fig. 2), and the region-2 fluorescence is collected by a photomultiplier tube (PMT) and demodulated in a lock-in amplifier. This demodulated fluorescence composes the data shown in Fig. 3(a).

The broad feature in these data is the 10-MHz-wide absorption line shape corresponding to the $3 \rightarrow 2$ transition. The Raman transition appears

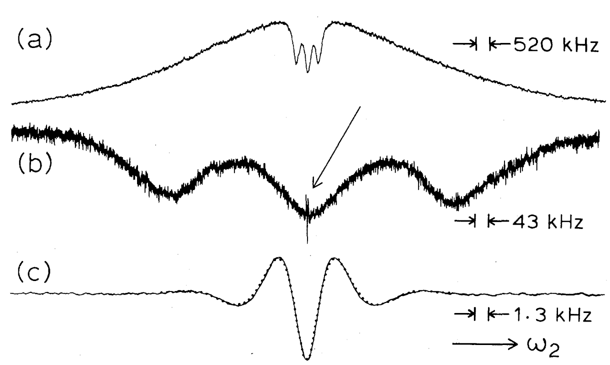

FIG. 3. (a) Raman dips on a 10-MHz-wide fluorescence background. (b) Expanded scan of the three Raman dips with Ramsey fringes corresponding to L = 15 cm on the central dip. (Arrow points to Ramsey fringes.) (c) Expanded scan of the Ramsey fringes with superimposed (dotted) theoretical curve. Scan rate: 110 Hz/sec; τ = 1 sec.

as three dips instead of only one in the center of this broad line shape. These three dips correspond to Raman transitions between different magnetic sublevels in the 1 and 3 states and are separated because we applied a 300-mG Zeeman field along the laser beam propagation direction. The central dip corresponds to the $m_F=0$, $\Delta m_F=0$ Raman transition and the two dips on either side correspond to the $m_F=\pm 1$, $\Delta m_F=0$ Raman transitions.

Figure 3(b) is the demodulated region-2 fluorescence obtained using the method of separated oscillatory fields with a field separation of $L=15$ cm. It is obtained by allowing both beams A and B to interact with the atomic beam. The three broad dips in Fig. 3(b) are the Raman transitions induced in region 2 and are the same as the three dips of Fig. 3(a), except for the expanded frequency scale. The extremely narrow features in the middle of the central $m_F=0$, $\Delta m_F=0$ Raman dip in Fig. 3(b) are the Ramsey fringes obtained only when both interaction regions, 1 and 2, are present.

An expanded scan of these Ramsey fringes appears in Fig. 3(c). In this case, background fluorescence is suppressed by chopping beam A instead of ω_1. Fringe symmetry is adjusted by varying the path length of beam A with respect to that of beam B for a given interaction-region separation, L. The width of the central fringe, 1.1 kHz (HWHM), is consistent with the $L=15$ cm interaction-region separation used in this case. It should be noted that no fringes appear on the outer $m_F=\pm 1$, $\Delta m_F=0$ Raman dips in Fig. 3(b) because they are washed out by stray ac magnetic fields present in the laboratory which do not affect the field-insensitive $m_F=0$, $\Delta m_F=0$ Raman transition. However, for a much smaller separation, $L=1$ cm, where the fringe width is 17 kHz (HWHM), we did observe Ramsey fringes associated with the $m_F=\pm 1$, $\Delta m_F=0$ Raman transitions and their amplitudes were the same as those corresponding to the $m_F=0$, $\Delta m_F=0$ transition.

The dotted curve superimposed on the data in Fig. 3(c) is the velocity-averaged fringe shape based on a preliminary calculation we have made for the conditions in our experiment. This calculated line shape, which is scaled vertically but otherwise involves no free parameters, is identical to that for a two-level atom in the strong-field limit.[4]

By increasing L to 30 cm we obtained narrower Ramsey fringes shown in Fig. 4(a). The measured width is 650 Hz (HWHM) which is again consistent with the transit-time width for $L=30$ cm. We should emphasize that the amplitudes of these fringes are the same as those for $L=1$ or 15 cm, thus demonstrating the absence of any significant line-broadening mechanisms other than that caused by transit time.

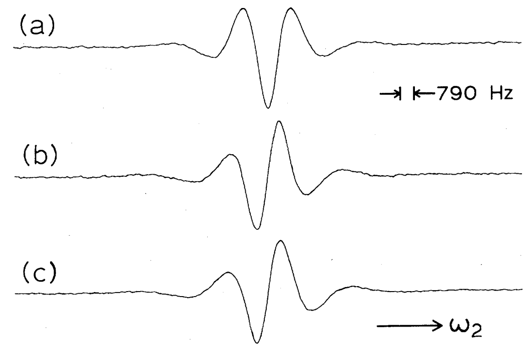

FIG. 4. Ramsey fringes corresponding to $L=30$ cm. Scan rate: 66 Hz/sec; $\tau=1$ sec. (a) Symmetric fringes with 650 Hz (HWHM) width. (b) Asymmetric fringes obtained with the path of beam A increased by 42 mm. (c) Discriminant obtained using frequency modulation.

Figure 4(b) shows the effect on the fringe line shape caused by varying the path traversed by beam A before interaction with the atomic beam. This trace is obtained with an increase in the path of beam A of 42 mm (i.e., $\frac{1}{4}$ of the equivalent microwave transition wavelength) over that used to obtain Fig. 4(a). In this case, the central fringe has a dispersive shape which again can be directly compared with that predicted by Ramsey for a two-level microwave transition.

We made a crude attempt to stabilize the frequency of the microwave oscillator using the discriminant shown in Fig. 4(c), corresponding to $L=30$ cm. This discriminant is obtained by removing the chopper from beam A and, instead, frequency modulating $\omega_1-\omega_2$ by a 2-kHz peak-to-peak excursion at a rate of 270 Hz. All other experimental conditions are identical to those used to obtain Fig. 4(a). The residual error signal obtained in this stabilization attempt was about 3 Hz peak to peak for a 1-sec integration time which gives $\Delta f/f \approx 2.5\times 10^{-10}$, $\tau=1$ sec. It should be noted that if cesium were used instead of sodium in the present apparatus the short-term stability would be $\Delta f/f \approx 2.5\times 10^{-11}$, $\tau=1$ sec for the same signal-to-noise ratio. This is because the cesium hyperfine splitting is about 4.5 times greater than that of sodium and because of the longer transit time due to the larger atomic mass of cesium. These preliminary data compare well with the short-term stability of a conventional cesium atomic beam clock for an interaction-region separation of 30 cm and averaging time of 1 sec.

We plan to investigate further the applicability of this stimulated, resonance Raman technique to frequency standards. Of particular concern are misalignment and second-order Doppler[9] effects, as well as level shifts due both to laser intensity variations and to the fluorescence from the interaction regions that propagates along the atomic beam.[12] In addition, we must consider errors due to ω_1 not being exactly on resonance with the $1 \rightarrow 2$ transition and also due to residual laser jitter on the order of the intermediate level lifetime. We will also attempt to enhance the signal-to-noise ratio by using higher oven temperatures and rectangular slits and perhaps employing multiple atomic beams. Detection efficiency can be increased by recycling the atoms via specialized optical pumping techniques which increase the number of scattered photons per atom.

We are particularly interested in extending this Raman technique into the millimeter region of the spectrum. For such large difference frequencies, the generation of the second laser frequency by modulation techniques becomes rather difficult. Therefore, for extension into the millimeter-wave and far-ir regions it will be necessary to use two independent lasers which are locked to a common Fabry-Perot cavity using wide-band stabilization techniques.[1] Atomic beam divergence will also be important at these frequencies but its effect can be minimized by using the three-zone interaction scheme first proposed by Baklanov, Dubetsky, and Chebotayev.[7]

To reduce the transit-time widths much further without using superlong interaction-region separations, it will be necessary to cool the atomic beam. Laser-cooling methods have already been studied[13] and are applicable in this case. Finally, the stimulated, resonance Raman technique may be directly applied to trapped ions[14] in which case Ramsey fringes may be obtained by using time-separated pulsed excitation.

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