PHYSICAL REVIEW A, VOLUME 61, 041404(R)

Laser modulation technique for single isotope spectroscopic studies

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(Received 20 December 1999; published 17 March 2000)

We demonstrate an atomic-beam approach to obtaining isotope specific driven-atom spectra using samples of arbitrary isotopic composition. The method employs modulation of a target isotope ground-state population by an upstream preparation laser. Desired driven-atom signals generated downstream from the preparation laser are isolated using a lock-in amplifier referenced to the preparation laser modulation frequency. In our experiments, an intensity-modulated laser resonant with the barium $(6s^2)^1S_0$-$(6s6p)^3P_1$ 791.1-nm intercombination transition selectively pumps ^{138}Ba atoms via radiative decays to the $(6s5d)^3D_{1,2}$ metastable states. Dressed-atom gain studies using the $(6s^2)^1S_0$-$(6s6p)^1P_1$ 553.5-nm transition demonstrate the efficacy of the method. This technique has general utility for selectively isolating specific isotopic signals in systems possessing optically controllable atomic-beam populations.

PACS number(s): 42.50.−p, 32.80.Bx, 32.30.−r, 31.30.Gs

Experimental quantum optics typically strives toward utilizing simple two- or three-level systems to provide unambiguous comparison between experiment and theory. Factors that tend to complicate experimental systems include level degeneracy, hyperfine splitting, and the presence of multiple, nearly degenerate isotopes. Atoms such as barium (Ba) have simple-level structures but still retain the complexity of a multi-isotope situation. We have developed a technique that isolates the signals of a single strongly driven isotope in the presence of multiple spectator isotopes.

To demonstrate the method, we employ an atomic beam of Ba. ^{138}Ba is the most abundant species [1,2]; however, the other six stable isotopes comprise over 28% of total abundance and add complexity to observations intended to focus solely on the response of ^{138}Ba. Moreover, while the even isotopes have zero nuclear spin, the odd isotopes possess hyperfine structure that additionally complicates the system. Our approach to ^{138}Ba selection consists of two stages. First, in the preparation region, a laser modulates the ground-state population of ^{138}Ba through time-dependent transfer of the population to metastable states (population shelving). Subsequently, downstream in the interaction region, lock-in detection of the probe laser, synchronous with the preparation laser modulation frequency, enables selective observation of the ^{138}Ba signals.

Figure 1 schematically depicts the experiment. An atomic beam composed of natural Ba is created by an effusion oven with a 3.2 mm-diam nozzle. Separate heaters maintain the temperatures of the nozzle (850 °C) and reservoir (725 °C) regions. The beam is collimated to <12 mrad divergence by a 1.2 mm-diam aperture placed 380 mm away inside a chamber evacuated to 10^{-7} torr using a liquid-nitrogen-trapped diffusion pump. A mechanical chopper (C1) situated in the vacuum chamber enables physical chopping of the entire atomic beam at 191 Hz. The mechanical chopper was selectively employed as outlined below.

A titanium-sapphire laser (Ti:sapphire) with a linewidth of 1 MHz acts as the preparation laser. The Ti:sapphire laser is servolocked to the $(6s^2)^1S_0$-$(6s6p)^3P_1$ 791.1-nm ^{138}Ba intercombination transition via saturated absorption signals generated in a reference cell. After passing through a cylindrical expander, the preparation laser beam is oval in cross section (5 mm×15 mm) and intersects the atomic beam at right angles 50 mm upstream from the interaction region. Placed at the beam waist of the expander, a flywheel chopper (C2) modulates the preparation laser beam intensity by 100% at a ~1 kHz rate. The isotopic frequency shifts on the $^1S_0$-$^3P_1$ transition enable selective excitation of ^{138}Ba [3] and the transition is easily saturated. The power in the preparation beam was 30 mW; however, reduction by 10× had no effect on the measurements. As shown in Fig. 2, the large radiative branching from $(6s6p)^3P_1$ to the $(5s6d)^3D_{1,2}$ states facilitates modulation of the ground-state ^{138}Ba population. The longitudinal velocity distribution of the atomic beam limits the maximum preparation-beam modulation rate. Owing to this velocity spread, modulation in the interaction region is substantially washed out as soon as the modulation period of the preparation laser becomes comparable or shorter than the rms atomic transit time from the preparation to the interaction regions. With our present apparatus, the rms transit time is 100 μs.

We note that preparation pumping can also be performed on the $(6s^2)^1S_0$-$(6s6p)^1P_1$ 553.5-nm transition, since decay to the $(5s6d)^1D_2$ metastable state provides population shelving. However, off-resonant excitation of the $(6s^2)^1S_0$

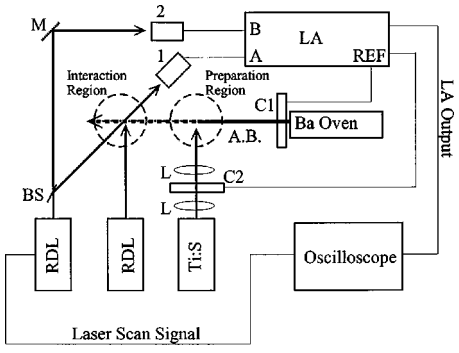

FIG. 1. A schematic representation of the experiment. RDL, ring dye laser; Ti:S, titanium-sapphire laser; M, mirror; BS, beam splitter; 1, 2, photodiodes; L, cylindrical lens: C1, mechanical chopper; C2, flywheel chopper; LA, lock-in amplifier.

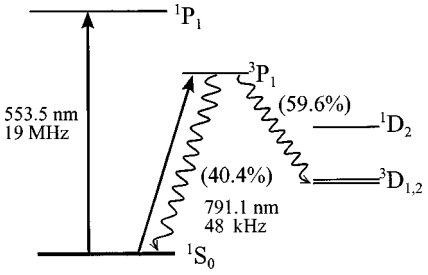

FIG. 2. A partial energy-level diagram for ^{138}Ba. Natural widths are given below the wavelengths. Branching ratios for the $^3P_1$ state are given along the indicated decay paths.

$(F=3/2)-(6s6p)^1P_1\,(F=5/2)$ transition of ^{137}Ba results in modulation of this isotope's population as well as that of ^{138}Ba owing to the large natural width on this transition and the relatively small isotope separation. A similar limitation on excitation contrast on the 553.5 nm transition was reported elsewhere [4].

Two ring dye lasers (RDL) provide a strong driving field nearly resonant with the $(6s^2)^1S_0$-$(6s6p)^1P_1$ 553.5-nm transition and a relatively weak probe field to measure absorptive features of the driven Ba atoms near the driven $^1S_0$-$^1P_1$ transition. Strong here implies having a Rabi frequency substantially greater than the $^1S_0$-$^1P_1$ transition natural width of 19 MHz [5]. The pump (probe) beam has a full-width-at-half-maximum (FWHM) diameter of 3.0 mm ($<$1.0 mm) and power of $\sim$100 mW (5 μW). Both beams have linewidths of $<$2 MHz, are linearly polarized in the same direction, intersect the atomic beam at right angles, and are angularly separated by 2.2°. Saturation spectroscopic techniques enable frequency locking of the strong pump field on or near the ^{138}Ba $(6s^2)^1S_0$-$(6s6p)^1P_1$ transition. Balanced detection removes intrinsic probe laser intensity noise. Overall system spectral resolution is $\sim$15 MHz.

Figure 3 shows the atomic-beam weak-field absorption spectrum as measured by tuning the probe laser with the strong-driving-field laser off. The modulated preparation laser is on (off) during acquisition of the solid (dashed) trace. ν_a is the ^{138}Ba $(6s^2)^1S_0$-$(6s6p)^1P_1$ resonance frequency and ν_p is the probe-field frequency. During measurement of the absorption spectrum with the preparation laser off, the entire atomic beam was mechanically chopped in order to maintain use of lock-in detection. The large dip in the dashed trace represents 34% single-pass absorption by ^{138}Ba, and the nearby features correspond to absorption by other isotopes. With the modulated preparation laser on and resonant with ^{138}Ba (solid line), absorption signals from the other isotopes do not appear.

Perhaps the most fundamental system in quantum optics is the dressed atom [6]. Representing the coupled quantum system comprised of a two-level atom and strong driving field, the energy eigenstates are composed of a ladder of doublets split by the generalized Rabi frequency and separated by the driving-field frequency. When probed with a weak field, population differences between dressed levels create gain and absorption features [7]. Remarkably, however, some gain features are actually inversionless in both the

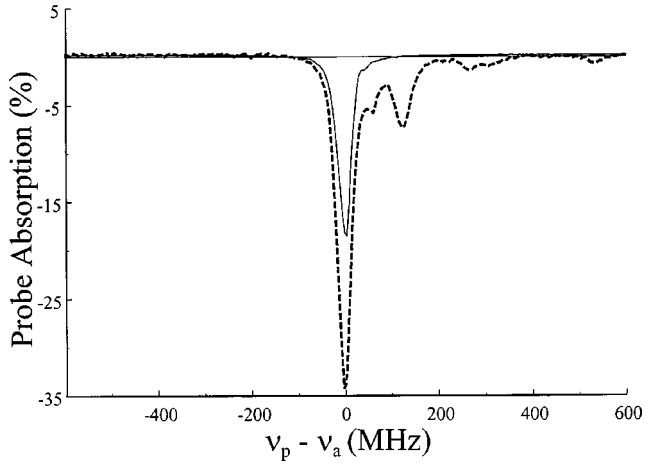

FIG. 3. Absorption spectrum of a weak tunable probe. Dashed (solid) line is the probe absorptive response to the full atomic (^{138}Ba only) beam. ν_p (ν_a) is the probe [^{138}Ba $(6s^2)^1S_0$-$(6s6p)^1P_1$ resonance] frequency.

atomic and dressed-state bases. Moreover, unlike the three-peaked fluorescence spectrum [8] whose general characteristics remain largely insensitive to the presence of multiple isotopes [9], the location, relative size, and shape of the dressed-atom absorption features are strongly dependent upon the driving-field detuning from the atomic resonance. Hence the presence of multiple isotopes greatly distorts the absorption spectrum away from that expected for a pure, single-isotope two-level system.

In Fig. 4, the solid (dashed) line depicts probe absorption in the presence of a strong, nonresonant driving field with (without) application of the ^{138}Ba-resonant preparation laser. In Fig. 4(a), $\Omega_G=255$ MHz and $\Delta=\nu_L-\nu_a=-170$ MHz, where ν_L is the pump driving-field frequency and Ω_G is the generalized Rabi frequency. The probe absorption spectrum obtained with the preparation laser active (solid trace) represents the classic driven two-level atom spectrum [7], i.e., a small gain peak ($\nu_p=\nu_L-\Omega_G$) as well as a large absorptive peak ($\nu_p=\nu_L+\Omega_G$) together with a small gain/loss dispersive feature (see magnified inset) near $\nu_P=\nu_L$. These features are precisely the signature of a driven two-level atom without the complicating signals from multiple isotopes seen in the dashed trace, demonstrating that the present experimental method effectively isolates the ^{138}Ba signal. Figure 4(b) shows the probe absorption spectrum obtained when the driving field is detuned toward a higher frequency, with $\Omega_G=245$ MHz and $\Delta=180$ MHz.

Figure 5 depicts probe absorption spectra obtained when the strong driving field is resonant with the ^{138}Ba $^1S_0$-$^1P_1$ transition. Figure 5(a) shows the probe absorption spectrum, with the modulated preparation laser on, producing a pure ^{138}Ba spectrum (solid trace) along with a theoretical spectrum (dotted trace) [7] representative of a two-level atom driven as per our experimental conditions with $\Omega_G=160$ MHz. Both gain (upward) and absorption features are observed. The gain produced is inversionless in both the bare and dressed-state bases. The smooth, symmetric spectrum

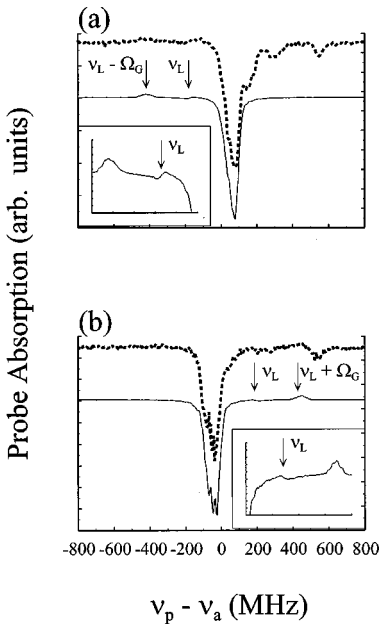

FIG. 4. Dressed-atom absorption spectrum with nonresonant pumping. Dashed (solid) line is probe absorptive response to the full atomic (^{138}Ba response) beam. ν_p (ν_a) is the probe [^{138}Ba $(6s^2)^1S_0$-$(6s6p)^1P_1$ resonance] frequency, ν_L is the pump-driving field frequency, Ω_G is the generalized Rabi frequency, and Δ the driving field detuning from ν_a. (a) $\Omega_G = 255$ MHz, $\Delta = -170$ MHz. (b) $\Omega_G = 245$ MHz, $\Delta = 180$ MHz.

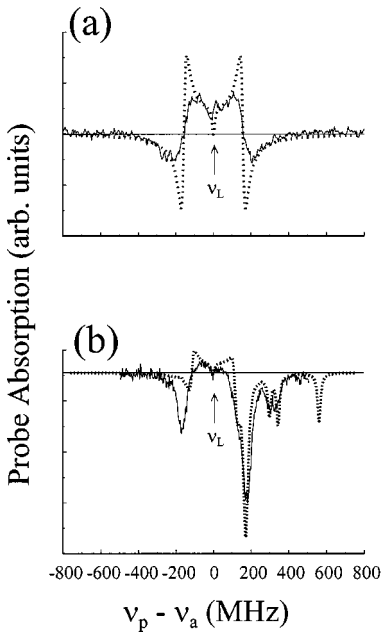

FIG. 5. Dressed-atom absorption spectrum with resonant pumping. The solid (dotted) line represents the experimental (theoretical) probe absorptive response. ν_p (ν_a) is the probe [^{138}Ba $(6s^2)^1S_0$-$(6s6p)^1P_1$ resonance] frequency, ν_L is the pump-driving field frequency, and Ω_G is the generalized Rabi frequency. (a) ^{138}Ba response, $\Omega_G = 160$ MHz. (b) Full atomic-beam response, $\Omega_G = 115$ MHz.

agrees remarkably well with the theory. This is contrasted with the probe absorption spectrum obtained with the preparation laser turned off in Fig. 5(b), where $\Omega_G = 115$ MHz. The presence of the isotopes grossly distorts the spectrum. Simulating the system as a sum over multiple, detuned two-level atoms representing the various isotopes and using appropriate weighting, gives fair agreement (dotted line). The anomalously large absorption observed at the lower frequency sideband demonstrates the limits of this simple treatment.

In conclusion, we have demonstrated a powerful yet simple technique that provides for the measurement of driven-atomic signals specific to a single isotope species, even in the presence of other isotopes. As such, this experimental technique provides an essential tool in the effort to provide rigorous and quantitative comparison of experiment and theory. Observations of dressed-state gain features demonstrate the efficacy of this approach. We note this technique also allows observation of fluorescence spectra of selected isotope species.

The authors gratefully acknowledge financial support from the National Science Foundation under Grant Nos. PHY-9421069 and PHY-9870223.

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