Forbidden Transitions in a Magneto-Optical Trap

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We report the first observation of a nondipole transition in an ultracold atomic vapor. We excite the $3P$-$4P$ electric quadrupole ($E2$) transition in ^{23}Na confined in a magneto-optical trap, and we demonstrate its application to high-resolution spectroscopy by making the first measurement of the hyperfine structure of the $4P_{1/2}$ level and extracting the magnetic dipole constant $A = 30.6 \pm 0.1$ MHz. We use cw optical-optical double resonance accompanied by photoionization to probe the transition.

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One of the frontiers in atomic physics is the detection of the signature of a transition that is classified as "forbidden." Forbidden-transition spectroscopy now plays a central role in tests of fundamental symmetries of nature. A significant example is the study of parity nonconserving interactions which provides one of the best measurements of electroweak symmetry breaking ([1] and references therein). More generally, forbidden transitions have been experimentally studied [2] using a variety of techniques (such as electron impact and laser excitation), in a range of contexts (from nebular spectra to cold-ion frequency standards), and in a number of atoms, ions, and molecules. In the case of alkali atoms, the first observation of a forbidden transition dates back to the early days of quantum mechanics [3]. More recently, nondipole effects have been explored in Stark-induced interference [4], photoionization [5], n-wave mixing (NWM) [6], and collision-induced absorption [7]. In the particular case of sodium, the $3S$-$3D$ $E2$ transition moment has been measured using NWM [8] and the $3P$-$(5P, 4F)$ transitions were observed in optical-optical double resonance (OODR) [9], both via pulsed laser excitation.

Another frontier in atomic physics is laser cooling and trapping of atoms. The availability of ensembles of cold atoms has made accessible entirely new regimes of atomic behavior ranging from atom-optical effects [10] to the formation of a Bose-Einstein condensate [11]. Moreover, a cold vapor is a nearly ideal enabler for precision measurement applications such as metrology [12] and high-resolution spectroscopy [13]. In this Letter, we describe the first experimental observation of a forbidden atomic transition in a laser-cooled vapor, confined in a magneto-optical trap (MOT). Our experiment combines the Doppler-free nature of the MOT, and its specific optical pumping properties, with the high resolution afforded by cw lasers and the near-unity efficiency of ion detection. To illustrate the power of this approach, we demonstrate the electric quadrupolar nature of the $3P$-$4P$ transition and use it to analyze the hyperfine structure of the $4P_{1/2}$ level [14]. Measurements of hyperfine splittings are of interest because they are sensitive to electronic correlations and relativistic effects, providing a benchmark for testing the accuracy of many-body atomic structure calculations [15].

Our initial observations were made with a standard MOT [16] (Fig. 1) operating on the $D2$ transition. We refer to this as the $D2$MOT. Atoms held in the trap were probed with light tunable around 750 nm generated using an Ar^+-pumped Ti:Sa ring laser. While the trapping light excited atoms on the $D2$ ($3S_{1/2} \rightarrow 3P_{3/2}$) transition, the probe laser excited the $3P$-$4P$ $E2$ transition. Atoms populating the resulting doubly excited state were then ionized to S and D states in the continuum by either a trap or a probe photon. The resulting Na^+ ions were detected using a channel electron multiplier (CEM).

Our experiments were typically carried out using probe laser powers $\sim$100 mW focused down to a $\sim$100 μm diameter spot yielding an excitation intensity of $\sim 10^6$ mW/cm^2 (the MOT diameter is $\sim$ 300 μm). The intensity I_{sat} for saturating the $E2$ transition can be estimated in the semiclassical approximation by equating the

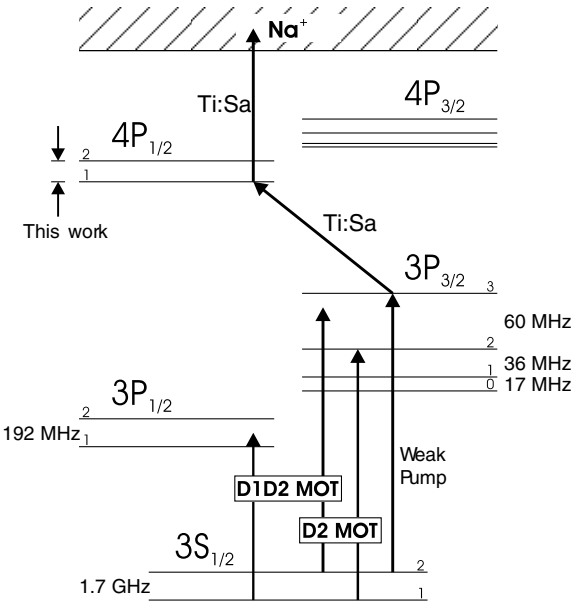

FIG. 1. Energy level diagram of sodium showing trapping configurations and OODR hyperfine splitting measurement.

$3P$-$4P$ quadrupole Rabi frequency ($\Omega_Q \sim QkE_o/\hbar$, where Q is the quadrupole moment of the transition, and k and E_o are the wave vector and electric field amplitude of the 750 nm light) with the sum of the linewidths of the $3P$ and $4P$ levels (γ_D), which are set by dipole ($E1$) emission to the $3S$ ground state. We find

$$I_{\text{sat}} = \frac{E_o^2}{2\eta_o} \sim \frac{1}{2\eta_o}\left(\frac{\hbar\gamma_D}{Qk}\right)^2, \quad (1)$$

where $\eta_o = 377\ \Omega$ is the impedance of free space. With $k = 2\pi/(750\text{ nm})$, $\gamma_D \sim 15$ MHz [17] and $Q \sim 30$ in atomic units [18] we arrive at $I_{\text{sat}} \sim 10^6$ mW/cm^2, indicating that the $E2$ transition is adequately saturated in our experiment. We were actually able to observe clearly resolved spectra with good signal-to-noise ratios down to ~20 mW of Ti:Sa power. It is important to note that saturation intensities comparable to those quoted here will also apply to transition moments to higher P states in Na [18] as well as in other alkalis [19]. This implies that our excitation techniques can be readily generalized to other levels and other species. Moreover, the low excitation power required means that similar $E2$ studies can be carried out even using inexpensive, low-power cw-diode lasers familiar to most cooling and trapping laboratories.

Our subsequent experiments were carried out using a MOT (Fig. 1), operating on both the $D1$ and $D2$ transitions. We refer to this trap as the $D1D2$MOT. In the $D1D2$MOT, the trapping light was produced by one dye laser detuned 16 MHz below the $3S_{1/2}(F = 2) \rightarrow 3P_{3/2}(F' = 3)$ $D2$ transition and locked using saturation spectroscopy. The repumping light was produced by a second dye laser that was intense enough to be resonant with both the 192 MHz split $3S_{1/2}(F = 1) \rightarrow 3P_{1/2}(F' = 1, 2)$ $D1$ transitions via power broadening. The power broadening eliminated the need to stabilize the repumping laser against long-term drift. Anti-Helmholtz coils produced a 20 G/cm magnetic field gradient and the background pressure was $\sim 10^{-9}$ Torr. From fluorescence measurements we estimated the number of atoms in the trap to be $\sim 10^6$ at densities $\sim 10^9$ cm^{-3}.

We find that the $D1D2$MOT is particularly useful for our experiments for several reasons. First, in the $D2$MOT, the presence of the repumping light tuned to the $3P_{3/2}$ excited-state manifold complicates the spectrum—the ion production channels involve two intense laser fields interacting with four closely spaced hyperfine levels. By moving the repumping frequency to the $3P_{1/2}$ manifold we isolate the $3P_{3/2}(F = 3) \rightarrow 4P_{1/2}(F' = 1, 2)$ spectrum and remove any effects associated with the repumper intensity. Second, in order to control power broadening in our spectrum and also to acquire data at detunings both above and below the $3P_{3/2}(F = 3)$ level, we switched the trapping light off and used a weak tunable pump to excite the $3S$-$3P$ transition. However, to prevent optical pumping into the $3S_{1/2}(F = 1)$ state from interfering with our measurement it was necessary to leave the repumper on, a configuration made possible by our two-laser scheme. Finally, the $D1D2$MOT naturally provides a steady-state population in the $3P_{1/2}$ state, allowing us to investigate all possible transitions between the $3P_J$ and $4P_{J'}$ fine structure levels, as discussed below.

Having observed the peaks in the ion spectrum associated with the hyperfine structure of the $4P_{1/2}$ level, we carried out several studies to confirm that excitation was due to an $E2$ process. To begin with, we noted that (i) the absence of a $J = 1/2 \rightarrow J' = 1/2$ peak in the $3P_J \rightarrow 4P_{J'}$ spectrum is a direct consequence of the ΔJ selection rules assuming an $E2$ process in an alkali atom [20] and (ii) the intensity scale agreement described in Eq. (1) above depends crucially on $E2$ excitation. We then eliminated the only two possible processes that could lead to excitation in the absence of an $E2$ process: Stark-mixing and collisional coupling. Consider first Stark mixing. In our setup, the only source of an electric field large enough ($\sim$ kV/cm) to produce Stark mixing is the CEM and associated ion collection optics. We switched off the CEM and still observed depletion of the MOT fluorescence at resonant Ti:Sa frequencies. While the presence of Stark mixing implies a quadratic dependence of the ion count on the electric field, no change was observed as the ion-detection-optic fields were varied by more than a factor of 4. To address the possibility of collisional effects, we repeated our measurements using the $D2$MOT in two distinct configurations that produce atom clouds differing in density by an order of magnitude. The first configuration, in which the trapping light was tuned below the $3P_{3/2}(F = 3)$ level, produced higher densities than the second configuration in which the trapping light was tuned below the $3P_{3/2}(F = 2)$ level. In the presence of collisional excitation, which scales as density squared, the ion-count rate is expected to change by approximately 2 orders of magnitude. Instead we observed an ion-count rate that increased only as the number of atoms in the interaction volume defined by the probe laser, or specifically, with a linear dependence on atomic density.

In our experiment we acquired spectra by varying the polarization of the 750 nm probe between linear, circular, and elliptical configurations and saw no measurable change in our signals. This is expected as the presence of an inhomogenous magnetic field in the volume of the MOT prevents the polarization of the 750 nm light from being well-defined in the interaction region. Hence the atomic ensemble is insensitive to changes in the polarization of the probe.

The quantitative measurement of the $4P_{1/2}$ hyperfine structure was made in the following manner (Fig. 1). The $D1D2$MOT trapping laser was passed through an acousto-optic modulator (AOM_{trap}) and the first order beam provided the trapping light. Using the modulator, the trapping light was switched on and off every 100 μs with a 50% duty cycle, which allowed us to maintain a trapped atom number of $\sim 10^5$. A weak ($\sim 1\ \mu\text{W/cm}^2$) pump was introduced into the trap by selecting a small

portion of light from the same laser before it passed through AOM_{trap} and sending it through a separate AOM whose first order output was tunable from 16 MHz below the $3P_{3/2}(F = 3)$ level to 64 MHz above. This pump field was not switched but was instead left on continuously. Further, the pump beam was retroreflected to prevent the MOT from being depleted due to mechanical pushing effects, particularly when the pump light was tuned near atomic resonance. In all experiments, repumping light was applied continuously.

Probe light from the Ti:Sa was also sent through a modulator ($AOM_{Ti:Sa}$) aligned such that the zeroth and first order beams had approximately equal power. The first order was shifted up in frequency relative to the zeroth order by 120.0045 ± 0.0003 MHz as measured on a precise frequency counter. Both orders were then separately focused on the atoms in the MOT and each beam contributed a set of peaks to our spectrum as the Ti:Sa frequency was varied (Fig. 2). The offset supplied by $AOM_{Ti:Sa}$ allowed us to calibrate our measurement and to check the slope and linearity of the Ti:Sa frequency sweep. We found the sweep was linear, but the nominal value of the slope required a 3% correction. This procedure eliminated any need to externally calibrate the Ti:Sa laser frequency.

Using this setup, we obtained ionization spectra for different detunings of the pump. Each spectrum was fit to the sum of four Lorentzians and a value of the hyperfine splitting was extracted from each fit. Forty-eight measurements were fit assuming a normal distribution. We thereby determined a mean value and standard error for the magnetic dipole constant $A(4P_{1/2}) = 30.6 \pm 0.1$ MHz, which is half of the hyperfine splitting. The theoretical prediction is 30.7 MHz [21], in excellent agreement with our data.

When we acquire spectra with the trapping light left on, we observe Autler-Townes doublets [22] which exhibit dependence on the Rabi frequency and detuning of the light. In the case of the $D1D2$MOT [Fig. 3(b)] we can identify these peaks as arising from the interaction of the intense trapping light with the $3P_{3/2}(F = 3)$ level. In the case of the $D2$MOT [Fig. 3(c)] the same interaction gives rise to all the peaks except for the rightmost which is due to the repumping light and which corresponds to the $3P_{3/2}(F = 2) \rightarrow 4P_{1/2}(F' = 2)$ transition. This type of spectrum reveals both the effective Rabi frequency seen by the atoms and the population distribution in the excited state of the $3P_{3/2}$ trapping manifold. The ability to detect these populations is of interest, for example, in elucidating the role of atomic hyperfine structure in excited-state cold collisions in MOTs [23]. In fact, $E2$ transitions are especially well-suited for this type of measurement for two reasons. First, the $\Delta F = 0, \pm 1, \pm 2$ selection rules allow for any single hyperfine level in any higher $P_{1/2}$ state to probe the entire trapping manifold. More generally, the weakness of $E2$ transitions makes them narrower probes than those that rely on dipole transitions from other atomic states. It is also interesting to note that, in the context of a MOT where exchange of momentum with light is important, the $E2$ transition is the lowest electric multipolar process in which exchange of orbital angular momentum between light beams and internal (as opposed to center-of-mass) atomic motion can be observed [24].

Having demonstrated that $E2$ excitation of an atomic transition can be observed using standard cw lasers in combination with a simple robust laser cooling apparatus, we now turn to extensions and limitations of our work. A natural first step is to consider the linewidths extracted from spectra such as in Fig. 2. These spectra, which were obtained after eliminating power broadening imply that the magnitudes of these linewidths are $\sim$10 MHz. Although these values confirm that the Na atoms are at or below the Doppler temperature ($\sim 240\ \mu$K) the observed linewidth is convolved with the natural linewidth of the

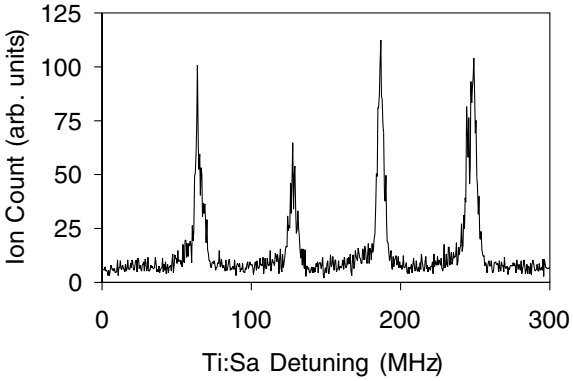

FIG. 2. Typical single scan ion spectrum of $4P_{1/2}$ hyperfine structure as a function of Ti:Sa frequency, acquired using the $D1D2$MOT. The first two peaks are due to the first order output of $AOM_{Ti:Sa}$ upshifted by $\sim$120 MHz from the zeroth order, which is responsible for the last two peaks. The origin of the abscissa is arbitrary.

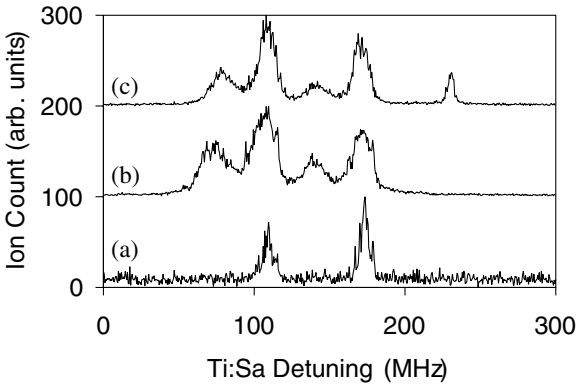

FIG. 3. Autler-Townes spectrum of $3P_{3/2}$ manifold probed from $4P_{1/2}(F = 1, 2)$ levels acquired with (a) a blue-detuned weak pump while the trapping light of the $D1D2$MOT is off, (b) the $D1D2$MOT, and (c) the $D2$MOT. The origin of the abscissa is arbitrary.

intermediate $3P_{3/2}(F = 3)$ state, which is known to be $\sim$10 MHz. For more accurate linewidth measurements, a detuning of the pump field further to the blue of the $3P_{3/2}(F = 3)$ level can be used because in this limit the spectrum yields the natural linewidth of the $4P$ state being ionized. Another way to measure the linewidth in this limit would be to detect the 330 nm fluorescence from the dipolar ($E1$) decay of the $4P$ level to the $3S$ ground state.

If we now consider the peak amplitudes in our spectra we find our experiment provides a natural way to measure the strength of the $E2$ interaction. For example, from the relative heights of the $4P_{1/2}$ peaks one could obtain the ratio of the quadrupole transition matrix elements for the $F = 1$ and 2 states. Similarly, one could compare the ion yield from the $3S \rightarrow 3P \rightarrow 4P \rightarrow$ continuum transition to that from the dipole-allowed process (which we observed with the probe laser at 820 nm) $3S \rightarrow 3P \rightarrow 3D \rightarrow$ continuum. The ratio of ion counts from the two processes essentially yields the value of the $E2$ transition matrix element in terms of the known $3P \rightarrow 3D$ matrix element [9]. We conclude from our experiment the possibility of measuring these matrix elements with better control over laser parameters.

The observation of a forbidden process in a physical system points to a lowering of symmetry, which in turn implies the presence of new physics. For example, our observations open up the possibility of using a MOT to observe second order ($\chi^{(2)}$) nonlinear processes which are not allowed in the dipole approximation in centrosymmetric media such as alkali vapors. Also interesting would be the observation of radiative behavior of cold atoms close to dielectric surfaces, a situation in which the intensity of quadrupole transitions has been predicted to become comparable to that of dipole transitions [25].

In conclusion, we report the first observation of a forbidden transition in a cold atomic vapor formed in a MOT. We use the resulting excitation process in combination with OODR ionization spectroscopy to measure the magnetic dipole constant of the $4P_{1/2}$ level of sodium and find $A = 30.6 \pm 0.1$ MHz. In addition to demonstrating the application to high-resolution atomic spectroscopy, we describe how our technique can be used to perform multiphoton ionization and Autler-Townes spectroscopy and to explore the role of hyperfine structure in alkali MOTs.

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