We report the first observation of a non-dipole transition in an ultra-cold atomic vapor. We excite the 3P-4P electric quadrupole (E2) transition in $^{23}$Na confined in a Magneto-Optical Trap (MOT), and 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 OODR (Optical-Optical Double Resonance) accompanied by photoinization 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 non-conserving (PNC) interactions which provides one of the best measurements of electro-weak symmetry breaking [1]. 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, non-dipole effects have been explored in photoionization [4], n-wave mixing (NWM) [5] and collision-induced absorption [6]. In the particular case of sodium, the 3S-3D E2 transition moment has been measured using NWM [7] and the 3P-(5P, 4F) transitions were observed in OODR [8], 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 [9] to the formation of a Bose-Einstein condensate [10]. Moreover, a cold vapor is a nearly ideal enabler for precision measurement applications such as metrology [11] and high-resolution spectroscopy [12]. In this Letter, we describe the first experimental observation of a forbidden atomic transition in a laser-cooled vapor, confined in a 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. 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 [13].

Our initial observations were made with a standard MOT [12] (Fig. 1) operating on the D2 transition. We refer to this as the D2MOT. 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 $\sim 100$ $\mu$m diameter spot yielding an excitation intensity of $\sim 10^6$ mW/cm$^2$ (the MOT diameter is $\sim 300$ $\mu$m). The intensity $I_{Sat}$ for saturating the E2 transition can be es-
timated in the semi-classical approximation by equating the 3P-4P quadrupole Rabi frequency ( $\Omega_Q \sim QKE_\circ/\hbar$ , where $Q$ is the quadrupole moment of the transition, and $k$ and $E_\circ$ are the wavevector and electric field amplitude of the 750 nm light ) with the sum of the linewidths of the 3P and 4P levels ($\gamma_D$ ), which are set by dipole (E1) emission to the 3S ground state. We find

$$I_{Sat} = \frac{E_\circ^2}{2\eta_o} \sim \frac{1}{2\eta_o} \left( \frac{\hbar \gamma_D}{Qk} \right)^2$$

where $\eta_o = 377 \Omega$ is the impedance of free space. With $k=2\pi/(750 \text{ nm})$, $\gamma_D \sim 15 \text{ MHz}$ and $Q \sim 30$ in atomic units [15], we arrive at $I_{Sat} \sim 10^6 \text{ mW/cm}^2$, 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 $\sim 20 \text{ mW}$ 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 [15] as well as in other alkalis [16]. 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 D1D2MOT. In the D1D2MOT, 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 $\sim 10^8$ at densities $\sim 10^9 \text{ cm}^{-3}$.

We find that the D1D2MOT is particularly useful for our experiments for several reasons. First, in the D2MOT, 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$4P$_{3/2}$($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$=1) state from interfering with our measurement it was necessary to leave the repumper on, a configuration made possible by our two-laser scheme. Finally, the D1D2MOT naturally provides a steady-state population in the 3P$_{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 (1) 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 $\Delta J$ selection rules assuming an E2 process in an alkali atom [15] and (2) 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 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 four. To address the possibility of collisional effects, we repeated our measurements using the D2MOT 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 two orders of magnitude [17]. 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.

The quantitative measurement of the 4P$_{1/2}$ hyperfine structure was made in the following manner (Fig. 4). The D1D2MOT 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$\mu$s with a 50% duty cycle, which allowed us to maintain a trapped atom number of $\sim 10^8$. A weak ($\sim 1 \text{ µW/cm}^2$) 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 16MHz below the 3P$_{3/2}$($F$=3) level to 64MHz above. This pump field was not switched, but was instead left on continuously. Further, the pump beam was retro-
reflect 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 $120.0045 \pm 0.0003$ 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 set-up, 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 [19], in excellent agreement with our data.

When we acquire spectra with the trapping light left on, we observe Autler-Townes doublets which exhibit dependence on the Rabi frequency and detuning of the light. In the case of the D1D2 MOT (Fig. 3a) 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. 3c) 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 spectra 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 [20]. 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 [21].

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). We observe that the magnitudes of these linewidths are $\sim10$ MHz. Although the values confirm that the Na atoms are at or below the Doppler temperature ($\sim240\mu$K) the observed linewidth is convolved with the width of the intermediate $3P_{3/2}(F=3)$ state, which is known to be $\sim10$ 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

![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 $\sim120$ MHz from the zeroth order, which is responsible for the last two peaks. The origin of the abscissa is arbitrary.](image)

![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, (c) the D2 MOT. The origin of the abscissa is arbitrary.](image)
the natural linewidth of the 4P state being ionized. An alternative approach 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. From the relative heights of the 4P levels we estimated the ratio of the quadrupole transition matrix elements for the F = 1 and 2 states to be 1. Similarly, we compared the ion yield from the 3S–3P→4P→continuum transition to that from the dipole-allowed process (with the probe laser at 820nm) 3S→3P→3D→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→3D matrix element Χ. Our experimental values agree with the theoretical value used to estimate Eq.(1) within a factor of 5 uncertainty. Better control over laser parameters can provide amplitude data of quality high enough to be compared to theory, and to be of use, for example, in PNC experiments. We point out that other methods for measuring forbidden transition matrix elements, such as NWM [2] and polarization spectroscopy [21] have so far been implemented using pulsed lasers. The application of cw techniques demonstrated by us would make these methods more accurate and increase their resolution enabling them to account for atomic hyperfine structure.

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 (χ(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 [22]. The achievement of single-atom MOTs [24] implies the possibility of PNC measurements via E1-E2 interference as proposed for Ba+ [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 ±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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[18] We observe the ion counts scale in this manner for the two MOTs when we produce highly excited Na[12].