A magic wavelength for an atomic transition is a wavelength for which the differential ac Stark shift vanishes [14]. The existence of magic wavelength enables independent control of internal hyperfine-spin and external center-of-mass motions of atoms (including atomic ions). Magic wavelengths provide extensive applications in quantum state engineering and precision frequency metrology [5]. It has been measured experimentally [3-6] in neutral atomic systems and demonstrated that the new generation of atomic clocks can be realized by utilizing neutral atoms in magic-wavelength optical lattices [1-12]. In this Letter, we report a novel application of the magic wavelength concept, namely the determination of the Ca\textsuperscript{+} 4s\textsubscript{1/2} → 4p\textsubscript{1/2} to 4s\textsubscript{1/2} → 4p\textsubscript{3/2} oscillator strength ratio to sub-0.5\% accuracy.

The precise knowledge of oscillator strengths is an important topic in atomic physics with one major application being their use as enabling data in astrophysical analysis [13]. Furthermore, recent advances in laser cooling technology has resulted in a number of new applications, such as degenerate gas [14], quantum information [15], and fundamental symmetry experiments [16]. Another motivation is the extreme precision achieved in the new generation of optical frequency standards which makes them sensitive to external electromagnetic perturbations. For example, atomic clocks at room temperature are sensitive to a radiation shift where the black-body radiation emitted by the apparatus enclosing the trap leads to differential Stark shifts in the energy of the two states of the clock transition [4, 10-12, 17-19]. Knowledge of the most important oscillator strengths, and thus the polarizabilities of the two states in the clock transition makes it possible to correct for black-body radiation shift, which also can be corrected by measuring differential scalar polarizability [21].

The experiment introduced here uses a probe laser to apply an ac Stark shift to the 40Ca\textsuperscript{+} 4s\textsubscript{1/2} → 3d\textsubscript{5/2} ion clock transition for the |m\textsubscript{j}| = 1/2 and |m\textsubscript{j}| = 3/2 magnetic sub-states of the 3d\textsubscript{5/2} level. Measurements of the ac Stark shifts at different frequencies allow the magic wavelengths near 395 nm, and thus the oscillator strength ratio to be determined. One advantage of the magic wavelength experiment is that it is effectively a null experiment. Therefore, it only requires that the intensity of the laser field to be stable and does not require precise knowledge of the intensity of the laser field.

The measured magic wavelengths near 395 nm lie between the resonant 4s\textsubscript{1/2} → 4p\textsubscript{1/2} and 4s\textsubscript{1/2} → 4p\textsubscript{3/2} transitions. Calculations had shown that these magic wavelengths were very sensitive to the ratio of the 4s\textsubscript{1/2} → 4p\textsubscript{3/2} to the 4s\textsubscript{1/2} → 4p\textsubscript{1/2} oscillator strengths [21]. The Ca\textsuperscript{+} resonant oscillator strength cannot be determined with a single measurement since the 4p\textsubscript{j} states can decay to either the 4s ground state or the 3d\textsubscript{J} excited states. Besides the 4p\textsubscript{j} lifetimes, one also needs the branching ratio for transitions to the 4s and 3d\textsubscript{J} states [22-24] since the 4p\textsubscript{j} → 3d\textsubscript{J} transitions makes a contribution of about 6\% to the lifetimes [21, 25, 26]. The advantage of the magic wavelength approach is that the polarizability contributions from the 4s\textsubscript{1/2} → 4p\textsubscript{j} transitions at the magic wavelength are three orders of magnitude larger than other contributions to the polarizabilities.

In some respects, the approach used to extract the oscillator strength ratio here is similar to that recently used
to estimate oscillator strength ratios from measurements of tune-out wavelengths \[27, 28\] for neutral potassium and rubidium \[29, 30\] (the dynamic polarizability of an atom in an ac field is zero at a tune-out wavelength). In both cases, the contributions to the polarizability from the interesting transitions are very much larger than the contributions from the background transitions.

For an ion in a single mode laser field, the energy shift of a given atomic state \(a\) can be written by \[2\]

\[
\Delta E_a = -\alpha_a(\omega) I + o(\tilde{F}^2)
\]

For the \(^{40}\text{Ca}^+\) optical clock, one of major experimental concerns is the frequency shift of the clock transition caused by electromagnetic radiation, which can be written as

\[
\hbar \Delta \nu = \Delta E_d(\omega) - \Delta E_s(\omega) = -\Delta \alpha(\omega) (I) + \Delta o(\tilde{F}^2)
\]

where \(\Delta \alpha(\omega)\) is the differential dipole-polarizability. \(\Delta \alpha(\omega)\) can be zero at the magic wavelength \(\lambda_m\) \((\lambda_m = c/\omega_m)\), where \(c\) is the speed of light in vacuum), and the residual term, \(\Delta o(\tilde{F}^2)\) is several orders of magnitude smaller than \(\Delta \alpha(\omega)\) and can be ignored. Using ion optical clock techniques, the differential light shift \(\Delta \nu\) can be measured accurately and so \(\lambda_m\) can be determined with high precision.

A sketch of the experimental setup for measurement of magic wavelengths is shown in Fig.1. The whole system is composed of two main parts, one is optical clock based on single trapped \(^{40}\text{Ca}^+\), and the other one is the 395 nm laser (L\(_m\)) system for measuring the light shift of the clock transition.

The detail of the optical clock has been described in previous work \[31, 33\]. A 729 nm probe laser was firstly locked to an ultra-stable, high finesse cavity mounted on a vibration isolation platform (TS-140) by the Pound-Drever-Hall method, and an acousto-optic modulator is used to cancel the slow linear drift of the reference cavity.

The L\(_m\) laser used in the experiment is frequency stabilized using a transfer cavity referenced to the 729 nm probe laser, and the long-term drift is reduced to less than 10 MHz within 4 hours. An unpolarized beam splitter (BS) is used to split a part of light for monitoring the laser power, which is 700 \(\mu\)W with a jitter of 3 \(\mu\)W. The power meter used in the experiment is a commercial power meter (S120VC, Thorlabs Inc.). The power of the incident and output beams of L\(_m\) laser are monitored simultaneously, and they agree with each other very well on the jitter of laser power. The power of L\(_m\) laser into the trap is 731(4) \(\mu\)W and the waist radius of the beam is 203(5) \(\mu\)m during the measurement. A polarized beam splitter is placed in the light path before the ion-light interaction maintains the linear polarization of L\(_m\) laser used to measure the magic wavelength. In this way, the linear polarization purity can reach 99.9\%, which can be derived by analyzing the polarization of the incident light and the transmission light of L\(_m\) laser.

In our work, a single \(^{40}\text{Ca}^+\) ion is trapped in a miniature Paul trap, and the ion is laser cooled to few mK. The single ion’s excess micromotion is minimized by adjusting the voltages of two compensation electrodes and two end-cap electrodes with the RF-photon correlation technique \[34\] before any measurements. The clock transition \((s-d)\) splits symmetrically into ten Zeeman components around the zero-field line center \[32\]. Then the probe laser is further referenced to the \(^{40}\text{Ca}^+\) ion clock transitions by feeding back to the frequency of the acousto-optic modulator (AOM1) to compensate for changes of the magnetic field and address of individual Zeeman transitions. In the experiment, the pulse sequences of 397 nm, 866 nm, 854 nm, and 729 nm lasers are similar to that used in \(^{40}\text{Ca}^+\) ion optical frequency standard \[32\]. The pulse sequence of the L\(_m\) laser is introduced to measure the light shift. The L\(_m\) laser is off during the Doppler cooling period, and is on and off alternately during probing stage to measure the light shift. The frequency values of AOM1 are recorded automatically every cycle by PC and the light shift caused by the L\(_m\) laser beam can be measured by calculating the difference of two cycles with the L\(_m\) laser on and off.

Ac Stark shifts within 0.2 nm around the magic wavelength \(\lambda_m\) was studied. Six randomly wavelengths of the L\(_m\) laser were chosen and the ac Stark shift was measured at each wavelength by switching on/off the L\(_m\) laser. The incident power of L\(_m\) laser on the ion probably change when tuning the wavelength, since the laser beam direction maybe change a little bit. Therefore the power was calibrated to ensure that it is identical within 4% at six randomly wavelengths. The measured six points are fitted linearly and the magic wavelength is obtained. Fig.2 shows one measurement of \(\lambda_{|m|=1/2}\).

Due to the influence of the uncertainties from wave-meter measurement, the broadband spectral component and the power jitter of L\(_m\) laser, it is difficult to obtain the frequency difference by separate measurements of \(|m_j| = 1/2\) and \(|m_j| = 3/2\). Here a new measurement protocol is adopted. In the experiment, the ac Stark shifts under the condition of \(|m_j| = 1/2\) and \(|m_j| = 3/2\) of 3\(d_5/2\) state at each one of the six wave-
lengths of $L_m$ laser were measured alternately. The $L_m$ laser’s wavelength is tuned from 395.7 nm to 395.9 nm, then from 395.9 nm to 395.7 nm. 10 times of $\lambda_{|m_j|=1/2}$, $\lambda_{|m_j|=3/2}$ measurements and the trimmed means, giving $\lambda_{|m_j|=1/2} = 395.7992(2)$ nm and $\lambda_{|m_j|=3/2} = 395.7990(2)$ nm, are presented in Fig. 3. The different value between $\lambda_{|m_j|=1/2}$ and $\lambda_{|m_j|=3/2}$ is 0.0002(6) nm, which agrees with the theoretical calculation [21].

To get the final magic wavelength measurement of the $L_m$ laser, systematic shifts must be considered and corrections should be applied to the above averaged frequencies. These shifts include the broad spectral component, the light polarization, the second order Doppler shift, the calibration of the wavemeter, etc. An error budget is given in Table I.

One of the most major errors of the magic wavelength measurement comes from the broad spectral component of $L_m$. To evaluate the broad spectral component, a grating spectrometer (IHR550, HORIBA) is used to analyze the laser spectrum and we found that >99% of the laser power is within the wavelength range of 0.03 nm, so only <1% laser power is out of that range, which corresponding <0.0005 nm contribution to uncertainty in $\lambda_{|m_j|=1/2}$ and $\lambda_{|m_j|=3/2}$ by theoretical calculation [30]. After that the spectral component in the range of 0.03 nm around the carrier was analyzed by observing the beatnote of the $L_m$ lasers with another similar laser using a spectrum analyzer. From the spectrum it mainly includes three components from which the ac Stark shift can be estimated on the base of proportion of three spectral components, and only an uncertainty of <0.0001 nm is obtained. Lights with different polarizations can result in different ac Stark shifts. Elliptical polarization will have both vector and scalar light shifts for atoms with $|m_F| > 0$ [32], changing the value of magic wavelength. In our experiment, a polarized beam splitter (PBS) is used to create a pure linear polarization, and ellipticity component is reduced to <0.1% by analyzing the light beam before and after the vacuum chamber. To evaluate the contribution due to non-linearly polarized component, the uncertainty with circularly polarized light was measured and the wavelength difference with linearly and circularly polarized light is <0.01 nm, thus there would be <0.0001 nm of uncertainty with <0.1% ellipticity.

The $L_m$ laser could heat the ion or deflect the efficiency of the laser cooling, introducing a second order Doppler shift and Stark shift due to the increase of the ion thermal motion or micromotion. To estimate the second order Doppler shift, the ion temperature is measured by serving the intensity of secular sidebands together with the measurements of the micromotion with rf-photon correlation method [34], with and without the $L_m$ laser. The $L_m$ laser wavelength after frequency stabilization is monitored by a wavemeter (High Finesse WS-7) with an absolute accuracy of 60 MHz after the calibration using the clock laser. Thus the total uncertainty is within 0.00006 nm.

### Table I: The magic wavelength measurement uncertainty budget.

| Source of uncertainty | Shift (pm) | Uncertainty (pm) |
|-----------------------|------------|------------------|
| Broadband light       | 0          | 0.60             |
| Light polarization    | 0          | 0.01             |
| Second order Doppler shift and Stark shift | 0.01 | 0.01 |
| Laser wavelength      | 0          | 0.06             |
| Statistical uncertainty | -     | 0.20             |
| **Total**              | **0.01**   | **0.7**          |

![FIG. 2: (color online) The ac Stark shifts at different laser wavelengths. Each point represents 2000 s of experimental data. The blue solid line is the linear fit to the data. The zero ac Stark shift wavelength is identified as $\lambda_{m_j}$. The inserted figure is magnification of one measurement point.](image)

![FIG. 3: (color online) (a) The 10 $\lambda_{|m_j|=1/2}$ measurements (black squares). (b) The 10 $\lambda_{|m_j|=3/2}$ measurements (blue triangle) under the same conditions with the $\lambda_{|m_j|=1/2}$. The solid red circle and the solid purple triangle indicate the trimmed mean and the errors are statistical errors of 10 $\lambda_{|m_j|=1/2}$ and $\lambda_{|m_j|=3/2}$ measurements.](image)
Two magic wavelengths, 395.7992(7) nm and 395.7990(7) nm in the spin-orbit energy gap of the 4p state were identified. The dynamic Stark shift is strongly dominated by the large and opposite polarizability contributions from the 4p_{1/2} and 4p_{3/2} states \cite{19,22,23}. The contributions of the 3d_{5/2} polarizabilities are typically small in magnitude at this wavelength. The dynamic Stark shift can be written as

\[ 0 = \alpha_{4s_{1/2}}(\omega_M) - \alpha_{3d_{5/2}}(\omega_M) \]

\[ \cong \frac{f(4s_{1/2} - 4p_{1/2})}{\epsilon^2_{4s_{1/2} - 4p_{1/2}} - \omega^2_M} + \frac{f(4s_{1/2} - 4p_{3/2})}{\epsilon^2_{4s_{1/2} - 4p_{3/2}} - \omega^2_M} + \Delta \]

where \( \Delta \) consists of the remainder terms in the 4s_{1/2} polarizability and the 3d_{5/2} polarizability and is small with estimates of 2.95 \( a_0^3 \) for 3d_{5/2}=-1/2 state and 0.31 \( a_0^3 \) for the 3d_{5/2}=3/2 state \cite{21}. The value of \( f(4s_{1/2} \rightarrow 4p_{1/2}) \) was set to 0.3171 \cite{21}.

The 4s_{1/2} \rightarrow 4p_{3/2;1/2} oscillator strength ratio, \( R_f \), is

\[ R_f = \frac{f(4s_{1/2} - 4p_{3/2})}{f(4s_{1/2} - 4p_{1/2})} = 2.029(5) \] (4)

The line strength ratio, \( R_s \) is

\[ R_s = \frac{|\langle 4s||D||4p_{3/2}\rangle|^2}{|\langle 4s||D||4p_{1/2}\rangle|^2} = 2.011(6) \] (5)

A change in \( \Delta \) of 2.0 \( a_0^3 \) will result in the derived \( R_s \) changing by 0.0010. Changes in the oscillator strengths of the background transitions of more than 5% would be needed to change \( \Delta \) by 2.0 \( a_0^3 \) and the uncertainty estimates in \( R_s \) and \( R_f \) allows for this. Previous estimates of the line strength ratio have been made using the relativistic all-order many-body perturbation theory giving 2.001 \cite{20} and relativistic semi-empirical potential giving 2.0014 \cite{21}.

In summary, experimental determinations of the magic wavelengths of the \( ^{40}\mathrm{Ca}^+ \) clock transition are made with uncertainty better than 0.001 nm, which is firstly realized in the ion optical clock systems. The specific values are \( \lambda_{m,j=1/2} = 395.7992(7) \) nm and \( \lambda_{m,j=3/2} = 395.7990(7) \) nm. The 4s_{1/2} \rightarrow 4p_{3/2;1/2} line strength ratio is 2.011(6). The uncertainty from broadband light and statistical error were the largest contributors to the total uncertainty. A cavity for mode selection to the \( L_m \) laser can be used to reduce the uncertainty from broadband light and the uncertainty from statistical error can be improved by improving power stabilization. An order of magnitude improvement in the precision of these measured magic wavelengths is achievable. The method of Magic wavelength measurements can be applied on other single ion optical clock systems, such as \( \mathrm{Sr}^+ \) and \( \mathrm{Yb}^+ \). It has also been proposed that all-optical trapping of ions offers new possibilities in the simulation of quantum spin systems, ultracold chemistry with ions and more \cite{40,41}. It is possible that one can trap ions with the laser at the magic wavelength, which is hopeful for reducing the uncertainty both in the optical clock and the precision spectroscopy.

Acknowledgments

We thank T. shi, L. Tang for valuable suggestion. Thank C. Lee, X. Guan, Z. Yan, J. Ye and H. Klein for fruitful discussions. And thank X. Huang, H. Shu, H. Fan, B. Guo, Q. Liu, W. Qu, J. Cao, and B. Ou for the early experiments. This work is supported by the National Basic Research Program of China (2012CB821301), the National Natural Science Foundation of China (11474318, 91336211 and 11034009) and Chinese Academy of Sciences.

\begin{thebibliography}{10}

\bibitem{1} M. Takamoto, F.-L. Hong, R. Higashi, and H. Katori, Nature 345, 321 (2005).
\bibitem{2} Z. W. Barber, J. E. Stalnaker, N. D. Lemke, N. Poli, C. W. Oates, T. M. Fortier, S. A. Diddams, L. Hollberg, C. W. Huyt, A. V. Taichenachev, et al., Phys. Rev. Lett. 100, 103002 (2008).
\bibitem{3} L. Yi, S. Mejri, J. J. McFerran, Y. Le Coq, and S. Bize, Phys. Rev. Lett. 106, 073005 (2011).
\bibitem{4} J. Mitroy, M. S. Safronova, and C. W. Clark, J. Phys. B 43, 202001 (2010).
\bibitem{5} J. Ye, H. J. Kimble, and H. Katori, Science 320, 1734 (2008).
\bibitem{6} A. Brusch, R. Le Targat, X. Baillard, M. Fouché, and P. Lemonde, Phys. Rev. Lett. 96, 103003 (2006).
\bibitem{7} M. Takamoto, H. Katori, S. I. Marmo, V. D. Ovsianikov, and V. G. Pal’chikov, Phys. Rev. Lett. 102, 063002 (2009).
\bibitem{8} N. D. Lemke, A. D. Ludlow, Z. W. Barber, T. M. Fortier, S. A. Diddams, Y. Jiang, S. R. Jefferts, T. P. Heavner, T. E. Parker, and C. W. Oates, Phys. Rev. Lett. 103, 063001 (2009).
\bibitem{9} A. D. Ludlow, T. Zelevinsky, G. K. Campbell, S. Blatt, M. M. Boyd, M. H. G. de Miranda, M. J. Martin, J. W. Thomsen, S. M. Foreman, J. Ye, et al., Science 319, 1805 (2008).
\bibitem{10} B. J. Bloom, T. L. Nicholson, J. R. Williams, S. L. Campbell, M. Bishof, X. Zhang, W. Zhang, S. L. Bromley, and J. Ye, Nature 506, 71 (2014).
\bibitem{11} H. S. Margolis, J. Phys. B 42, 154017 (2009).
\bibitem{12} N. Hinkley, J. A. Sherman, N. B. Phillips, M. Schioppo, N. D. Lemke, K. Beloy, M. Pizzocaro, C. W. Oates, and A. D. Ludlow, Science 324, 1215 (2013).
\bibitem{13} R. L. Kurucz, Can. J. Phys. 89, 417 (2011).
\bibitem{14} M. S. Safronova, U. I. Safronova, and C. W. Clark, Phys. Rev. A 86, 042505 (2012).
\bibitem{15} A. V. Gorshkov, A. M. Rey, A. J. Daley, M. M. Boyd, J. Ye, P. Zoller, and M. D. Lukin, Phys. Rev. Lett. 102, 110503 (2009).
\end{thebibliography}
[16] A. A. Vasilyev, I. M. Savukov, M. S. Safronova, and H. G. Berry, Phys. Rev. A 66, 020101 (2002).
[17] P. Gill, Royal Soc. of London Phil. Trans. Series A 369, 4109 (2011).
[18] K. Beloy, J. A. Sherman, N. D. Lemke, N. Hinkley, C. W. Oates, and A. D. Ludlow, Phys. Rev. A 86, 051404 (2012).
[19] T. Rosenband, D. B. Hume, P. O. Schmidt, C. W. Chou, A. Brusch, L. Lorini, W. H. Oskay, R. E. Drullinger, T. M. Fortier, J. E. Stalnaker, et al., Science 319, 1808 (2008).
[20] P. Dubé, A. A. Madej, M. Tibbo, and J. E. Bernard, Phys. Rev. Lett. 112, 173002 (2014).
[21] Y.-B. Tang, H.-X. Qiao, T.-Y. Shi, and J. Mitroy, Phys. Rev. A 87, 042517 (2013).
[22] A. Gallagher, Phys. Rev. 157, 24 (1967).
[23] R. N. Gosselin, E. H. Pinnington, and W. Ansbacher, Phys. Rev. A 38, 4887 (1988).
[24] J. Jin and D. A. Church, Phys. Rev. Lett. 70, 3213 (1993).
[25] R. Gerritsma, G. Kirchmair, F. Zähringer, J. Benhelm, R. Blatt, and C. F. Roos, Eur. Phys. J. D 50, 13 (2008).
[26] M. S. Safronova and U. I. Safronova, Phys. Rev. A 83, 012503 (2011).
[27] L. J. LeBlanc and J. H. Thywissen, Phys. Rev. A 75, 053612 (2007).
[28] T. Topcu and A. Derevianko, Phys. Rev. A 88, 053406 (2013).
[29] C. D. Herold, V. D. Vaidya, X. Li, S. L. Rolston, J. V. Porto, and M. S. Safronova, Phys. Rev. Lett. 109, 243003 (2012).
[30] W. F. Holmgren, R. Trubko, I. Hromada, and A. D. Cronin, Phys. Rev. Lett. 109, 243004 (2012).
[31] Y. Huang, Q. Liu, J. Cao, B. Ou, P. Liu, H. Guan, X. Huang, and K. Gao, Phys. Rev. A 84, 053841 (2011).
[32] Y. Huang, J. Cao, P. Liu, K. Liang, B. Ou, H. Guan, X. Huang, T. Li, and K. Gao, Phys. Rev. A 85, 030503 (2012).
[33] H. Guan, Q. Liu, Y. Huang, B. Guo, W. Qu, J. Cao, G. Huang, X. Huang, and K. Gao, Optics Commun. 284, 217 (2011).
[34] D. J. Berkeland, J. D. Miller, J. C. Bergquist, W. M. Itano, and D. J. Wineland, J. Appl. Phys. 83, 5025 (1998).
[35] I. H. Deutsch and P. S. Jessen, Optics Commun. 283, 681 (2010).
[36] B. Arora, M. S. Safronova, and C. W. Clark, Phys. Rev. A 76, 052509 (2007).
[37] A. A. Madej, P. Dubé, Z. Zhou, J. E. Bernard, and M. Gertsvolf, Phys. Rev. Lett. 109, 203002 (2012).
[38] G. P. Barwood, G. Huang, H. A. Klein, L. A. M. Johnson, S. A. King, H. S. Margolis, K. Szymaniec, and P. Gill, Phys. Rev. A 89, 050501(R) (2014).
[39] N. Huntemann, M. Okhapkin, B. Lipphardt, S. Weyers, C. Tamm, and E. Peik, Phys. Rev. Lett. 108, 090801 (2012).
[40] I. V. Krasnov and L. P. Kamenshchikov, Optics Commun. 312, 192 (2014).
[41] M. Enderlein, T. Huber, C. Schneider, and T. Schaetz, Phys. Rev. Lett. 109, 233004 (2012).