Photoassociation of Laser-cooled Ytterbium Atoms

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Abstract
We report photoassociation of laser-cooled ytterbium (Yb) atoms. By detecting the trap loss of $^{174}$Yb atoms in the FORT due to the photoassociation beam, we could observe more than 90 photoassociation resonances of vibrational levels in the $^1\Sigma_u^+$ state which connects asymptotically to the $^1S_0 + ^1P_1$ atomic state in the dissociation limit. From the observed resonance frequencies we could precisely determine the atomic radiative lifetime of the $^1P_1$ state to 5.464 ± 0.005 ns. We have also observed linebroadening of photoassociation resonances, which is ascribed to the predissociation to the triplet states, and determined the transition probability to be 0.2. Furthermore, we have observed the decrease of the intensity of photoassociation signal at 435 GHz detuning from the $^1S_0 + ^1P_1$ asymptote, from which the scattering length is estimated to be equal to or less than 3 nm.

1 Introduction
Recently there has been increasing interest in cooling alkaline-earth-like atoms. There are two kinds of transitions from the ground state which can be used for cooling, i.e., the strong dipole-allowed singlet transition $^1S_0 \leftrightarrow ^1P_1$, and the weakly allowed intercombination transition $^1S_0 \leftrightarrow ^3P_1$. Although the cooling using the singlet transition can provide strong radiation pressure, the Doppler-cooling limit is on the order of mK, and the sub-Doppler cooling mechanism does not work for the bosonic isotopes which have no nuclear spin. In addition, the large branching from the $^1P_1$ state to the lower singlet or triplet states severely limited the lifetime of the magneto-optical trap (MOT). On the contrary, the cooling with the intercombination transition has many advantages. Since the radiative lifetime ranges from about ms for light atoms like Mg and Ca to about $\mu$s for heavy atoms like Ba and Yb, only the Doppler cooling mechanism is enough to cool down the atoms to $\mu$K or nK temperature region. In addition, it has no branching, and so the long lifetime of the MOT is expected. By overcoming some difficulties associated with the narrow linewidth of the intercombination transition, ultra-low temperatures of the alkaline-earth-like atoms were realized for Yb[1], $^{88}$Sr[2], and $^{40}$Ca[3] by the MOT using the intercombination transition.

Such low temperature is quite important to improve the performance of atomic clock using the alkaline-earth-like atoms. In addition, such new cooling scheme has opened new possibility of quantum degeneracy of alkaline-earth-like atoms. In fact, Ido et al., [4] have obtained very high phase space density of about 0.1 for the $^{88}$Sr atoms in a far-off-resonant-dipole trap (FORT). Recently we have also achieved similarly high phase-space density for $^{174}$Yb (boson) by a forced evaporative cooling in a crossed FORT. The sympathetic cooling effect has been also observed in the boson($^{174}$Yb)-fermion($^{171}$Yb) mixture in a crossed FORT, which leads to $T/T_F = 6$ where $T_F$ is the Fermi temperature[5].

To make further progress toward quantum degeneracy and atomic clock, infor-
Figure 1: Pair potential of Yb$_2$. The photoassociation light of about 399 nm excites the $^1S_0 + ^1S_0$ atoms to the molecular excited state $^1\Sigma_u^+$ which connects asymptotically to the $^1S_0 + ^1P_1$ atomic state in the dissociation limit. The long-range potential of the $^1\Sigma_u^+$ state is determined by the resonant dipole-dipole interaction $-C_3/R^3$.

Formation of interatomic interaction such as scattering length is of crucial importance. Since the photoassociation spectroscopy is a powerful method to obtain interatomic interaction, as has been demonstrated for alkali atoms[6], it is important to perform the photoassociation spectroscopy for alkaline-earth-like atoms. The first important photoassociation experiment among the alkaline-earth-like atoms was reported for $^{40}$Ca[7].

Here, we report the photoassociation spectroscopy of laser-cooled $^{174}$Yb atoms. By detecting the trap loss of $^{174}$Yb atoms in the FORT due to the photoassociation beam, we could observe more than 90 photoassociation resonances of vibrational levels in the $^1\Sigma_u^+$ state of Yb$_2$. This allows us to obtain useful information of interatomic interaction of $^{174}$Yb atoms such as predissociation and scattering length, as well as the accurate value of radiative lifetime of the $^1P_1$ state.

Figure 1 schematically shows the pair potential of $^{174}$Yb$_2$ molecule. In this work, we excited the transition from the $^1S_0 + ^1S_0$ atomic state to the excited molecular state $^1\Sigma_u^+$ which connects asymptotically to the $^1S_0 + ^1P_1$ atomic state in the dissociation limit. The long-range potential of the $^1\Sigma_u^+$ state is determined by the resonant dipole-dipole interaction $-C_3/R^3[8]$, where

$$ C_3 = \frac{3\hbar\lambda^3}{16\pi^3}. $$

Here, $\lambda$ is the transition wavelength of the $^1S_0 \leftrightarrow ^1P_1$ transition, and $\tau$ is the radiative
Figure 2: Experimental setup. The dense and cold Yb atoms were trapped in the FORT after the MOT with the intercombination transition. The light for the photoassociation which was generated by the frequency-doubled Ti:sapphire laser was applied to the atoms in the FORT. The number of the atoms remained after the irradiation of the photoassociation beam was measured by the absorption imaging method with the CCD.

The vibrational energy structure near the dissociation limit can be given by the semi-classical formula [9]

\[ E(v) = D - X_0(v_D - v)^6, \]

\[ X_0 = \frac{\hbar^6}{\mu^3 G_3^2} \left[ \frac{\Gamma(4/3)}{2\sqrt{2\pi}\Gamma(5/6)} \right]^6. \]

Here, \( D \) is the energy in the dissociation limit, \( \mu \) is the reduced mass, \( v \) is the vibrational quantum number, and \( v_D \) is the vibrational quantum number at the dissociation limit, which may take noninteger value. The advantageous feature of this transition is the lack of the fine- and hyperfine structures. This greatly simplifies the interpretation of the photoassociation data, which is in good contrast with the cases of alkali atoms [6] and metastable helium atoms[10].

2 Experimental Setup

Figure 2 shows the experimental setup. Firstly, the \(^{174}\)Yb atomic beam was decelerated by the Zeeman slower using the injection-locked GaN LD system[11]. The slowed \(^{174}\)Yb atoms were then trapped and cooled with the MOT with intercombination transition \(^1S_0 \leftrightarrow ^3P_1[1]\). The number of atoms was about \(10^8\), and the temperature was about 10 – 20\(\mu\)K, and the atom density was about \(10^{11} – 10^{12}/\text{cm}^3\). The atoms were then transferred to the FORT[12]. The beam for the FORT was generated by a frequency-doubled Nd\(^{3+}\)-YVO\(_4\) laser of 532 nm and 10 W, and was focused to the beam waist of 14\(\mu\)m, which gives the potential depth of about 1 mK. The number of atoms was about \(10^6\), and the temperature was about 100\(\mu\)K, and the atom density was as high as \(10^{13}/\text{cm}^3\). It is noted that the higher atom density and stability provided by the FORT was essentially important in our photoassociation experiment.
Figure 3: Photoassociation signal. The number of the atoms in the FORT indicated by cross was plotted against the detuning from the $^1S_0 + ^1P_1$ asymptote. The solid line is the Lorentzian fit. This resonance was assigned to the vibrational quantum number of $v_D' - v = 177$.

The 399-nm light for the photoassociation was generated by frequency-doubled Ti:sapphire laser, the frequency of which was offset-locked to the absolutely stabilized reference cavity and was measured by a wavemeter. The laser linewidth of about 1MHz was much smaller than the atomic linewidth of 29 MHz for the singlet transition $^1S_0 \leftrightarrow ^1P_1$. The photoassociation beam was applied for about 200ms during the FORT. We observed the photoassociation signal by measuring the trap loss due to the photoassociation beam, while scanning the Ti:sapphire laser frequency.

3 Experimental Results

Figure 3 shows a typical photoassociation signal of $^{171}$Yb atoms. This resonance was assigned to the vibrational quantum number of $v_D' - v = 177$, where $v_D'$ is the integer part of the $v_D$ corresponding to the highest vibrational quantum number. The signal was fitted with the Lorentzian curve with the width of 2.9 GHz. We could observe more than 90 photoassociation resonances of vibrational levels of the $^1\Sigma_u^+$ state. It is noted that none of the observed resonances exhibit apparent rotational structures. This comes from the fact that the expected rotational splitting is very small for heavy atoms like Yb, scaling with mass $M$ as $M^{-3}$. The rotational splitting of $J = 1$ and $J = 3$, for example, becomes only about 200 MHz for the $v_D' - v = 177$ resonance which has about 3 GHz linewidth. The lack of the rotational structure also comes from the fact that the temperature of 100 $\mu$K of the Yb atoms in the FORT was cold enough to exclude the rotational angular momentum other than zero for the colliding $^1S_0$ atoms due to the centrifugal potential barrier.
Figure 4: Resonance frequencies. The observed detuning with respect to the \(^1S_0 + ^1P_1\) asymptote indicated by cross was plotted against the assigned vibrational quantum number \(v'_D - v\). The solid line is the fit with the semiclassical formula of Eq. (2).

### 3.1 Lifetime of \(^1P_1\) state

Figure 4 shows the observed resonance frequencies where the detunings \(\Delta(v)\) from the \(^1S_0 + ^1P_1\) asymptote were plotted with the assigned vibrational quantum numbers \(v'_D - v\). The observed resonance positions \(\Delta(v) = (D - E(v))/h\) were fitted with Eq. (2). Here we did not include the rotational energy structures due to its smallness as mentioned above. We have obtained excellent fit of the Eq. (2) with the data where most of the deviations were less than 0.5%. From this fitting we could precisely determine the potential coefficient \(C_3\) from Eq. (3), and thus the atomic radiative lifetime of the \(^1P_1\) state from the Eq. (1) to be 5.464 ± 0.005 ns. This is about two-orders-of-magnitude improvement in accuracy compared with the previous measurements of level-crossings[13]. To further improve the fit, it would be helpful to include the higher dispersion terms such as \(C_6\) and \(C_8\).

### 3.2 Linebroadening

We have also observed linebroadening of photoassociation resonances. The linewidths were found to monotonously increase as the vibrational quantum numbers decreased. For example, the resonance corresponding to the \(v'_D - v = 83\) had the linewidth of about 100 MHz, and the resonance of \(v'_D - v = 148\) had 630 MHz linewidth, and the \(v'_D - v = 177\) resonance had 2.9 GHz linewidth.

We have investigated the origin of the observed linebroadening. The possibility of power-broadening was experimentally excluded. Furthermore, possible unresolved rotational structures are too small to explain the observed linebroadening, as was
discussed in the above. We believe that the predissociation from the $^{1}\Sigma _{u}^{+}$ state to the triplet molecular states is responsible for the observed broadening. The calculation of the Yb$_2$ molecular potential in Ref.[14] shows that the level-crossing occurs at an internuclear distance of about 6.5 Bohr radius between the $^{1}\Sigma _{u}^{+}$ state and the $^{3}\Pi _{u}$ state which connects asymptotically to the $^{1}S_0+^{3}P$ atomic states. See Fig. 1. The state-mixing between these states is caused by the spin-orbit interaction. Therefore, there is the channel of escape, i.e., predissociation, from the $^{1}\Sigma _{u}^{+}$ state to the $^{3}\Pi _{u}$ state. It is noted that the predissociation to the triplet molecular states which connect asymptotically to the $^{1}S_0+^{3}D$ atomic states would be also possible. The linewidth caused by the predissociation $\Gamma$ can be expressed by

$$\Gamma = (\Delta E_{v,v-1}/h) \times P,$$

where $\Delta E_{v,v-1}/h$ is the local vibrational splitting frequency, and $P$ is the Landau-Zener transition probability from the $^{1}\Sigma _{u}^{+}$ state to the triplet molecular states[15]. From our measurements, the probability $P$ was found to be about 0.2. This is compared with the theoretical calculation in Ref.[16] which predicts $P(^{1}\Sigma _{u}^{+} \rightarrow ^{3}\Pi _{u}) \sim 0.64$. The agreement between the theory and experiment on this level is impressive when we consider the many approximations in the calculation. It is interesting to directly detect the dissociating atoms which would be in the $^{3}P_2, ^{3}P_1, ^{3}P_0$ or $^{3}D_2, ^{3}D_1$ states, as in the experiment using potassium atoms[17]. This will finally fix the routes of the predissociation.

### 3.3 Intensity Modulation

Furthermore, we have observed the decrease of the intensity of photoassociation signal at 435 GHz detuning, which corresponds to the Condon point of 2.95 nm. Since the intensity of the photoassociation signal is proportional to the square of the ground-state wavefunction at the Condon point[6], the observed reduction of the intensity at 435 GHz detuning indicates that the ground-state wavefunction has the node at 2.95 nm. The ground-state wavefunction has its node also due to the attractive potential of $C_6$ at $R \leq R_Q = 1/2(mC_6/h^2)^{1/4}[6]$, where $m$ is the mass of $^{174}$Yb atom. The value of $C_6$ estimated from the London’s formula gives $R_Q$ of also about 3 nm. Thus we cannot definitely conclude that the scattering length is 2.95 nm. Instead, since we did not observe the decrease of the intensity at smaller detunings, we can conclude that the scattering length is equal to or less than about 3 nm. Although the investigation of the signal intensity at larger detunings is quite helpful for the discussion of the scattering length, the considerable linebroadenings made the observation of the signal at larger detunings quite difficult. The investigation of the vibrational series in the molecular ground state $^{1}\Sigma _{g}^{+}$ accessible with two-photon-photoassociation process would be necessary to undoubtedly determine the scattering length[6].

If the scattering length is 3 nm, the scaling law[18] tells us the successful generation of BEC of $^{174}$Yb by evaporative cooling in the crossed FORT, starting from the initial condition of $n \sim 10^{14}$ cm$^{-3}$ and $\rho \sim 10^{-4}$ already obtained in our experiment[12]. The effort toward this possibility is now underway.
4 Conclusion and Prospect

In conclusion, we have observed more than 90 photoassociation resonances of $^1\Sigma_u^+$ state of Yb. This allows us to obtain information of interatomic interaction of $^{174}$Yb atoms such as predissociation and scattering length, as well as the accurate value of radiative lifetime of $^1P_1$ state.

It is interesting to extend the photoassociation measurements to the isotopes other than $^{174}$Yb of the present work. The scattering length strongly depends on the vibrational quantum number at the dissociation energy $v_D$ which may well have different values between the isotopes. Possible large variation of the scattering length among the isotopes will be the great advantage of working with the Yb atom which features a rich variety of isotopes (five bosons and two fermions) and rather equally distributed natural abundances. One may tune isotope to obtain a different scattering length, instead of a magnetic field in the case of Feshbach resonance.

As another interesting future direction we propose the production of the Einstein-Podolsky-Rosen (EPR) pairs of cold fermionic $^{171}$Yb atoms with nuclear spin $I = 1/2$[19]. The Yb molecule of total nuclear spin $I = 0$ with well defined ro-vibrational state can be produced by the photoassociation of two cold $^{171}$Yb atoms, one of which is nuclear spin up and the other down. The successive (photo)dissociation from the molecule to the free atoms of opposite spin results in the generation of the EPR pairs. The high detection efficiency of atoms with the photo-ionization and ion-detection techniques allows us to test the Bell’s inequalities without loopholes using the EPR pairs of Yb atoms.

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References


