

Photoassociation Spectroscopy of Laser-Cooled Ytterbium Atoms

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We report the photoassociation spectroscopy of laser-cooled ytterbium atoms in an optical trap. We observed more than 90 photoassociation resonances of vibrational levels in the $^1\Sigma_u^+$ state, including 80 consecutive series, up to 490 GHz detuning with respect to the atomic resonance. From the resonance frequencies we derived the atomic radiative lifetime of the $(6s6p)^1P_1$ state to be 5.464 ± 0.005 ns, which is about 2 orders of magnitude improvement over previous results. We also observed line broadening of resonances, which is ascribed to the predissociation to the triplet states, and estimated the transition probability to be 0.2. Furthermore, we observed the decrease of the photoassociation signal intensity, from which the scattering length is estimated to be equal to or less than 3 nm.

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Photoassociation (PA), which was first proposed by Thorsheim *et al.* [1], is the phenomenon of optical formation of molecules from the free atoms, and has often been used for the accurate determination of the interatomic interaction including the scattering length. After the first observation of PA spectra using laser-cooled Na [2], PA spectroscopy has been reported for almost all cold alkali atoms [3], H [4], triplet He [5], and Ca [6].

Ytterbium (Yb) atom is a rare earth with electronic structure in the ground state of $(Xe)4f^{14}6s^2 (^1S_0)$ similar to the alkaline-earth atoms. Recently the high-density trapping of Yb atoms in a far-off resonant trap (FORT) [7] and the following achievement of Bose-Einstein condensation (BEC) of ^{174}Yb atoms [8] have been demonstrated, which is expected to be an important step for a future investigation of new phenomena. BEC in ultracold weakly interacting dilute gases is undoubtedly one of the most interesting research fields [9]. So far only BEC of the atoms with electron spins have been produced. Unlike the previously observed condensates, Yb BEC has distinct features connected to the two valence electrons, such as extremely narrow intercombination transitions which are ideal for future optical frequency standard [10], and insensitivity to external magnetic field which is important for precision coherent atom optics.

Quantitative description of BEC requires the knowledge of atom-atom collision data such as a scattering length. A scattering length of Yb atom has not been known even for the sign so far. Recently through the measurement of the chemical potential of the ^{174}Yb BEC, the scattering length of ^{174}Yb was estimated to be 6_{-5}^{+10} nm [8]. The relatively large uncertainty originated mainly from the inevitable uncertainty in the determination of the trapped atom density.

In this Letter, we report successful extension of the PA spectroscopy to this rare earth element, ^{174}Yb atoms. We detected the trap loss of atoms in a FORT due to the free $(^1S_0 + ^1S_0)$ -bound $(^1\Sigma_u^+)$ absorption induced by the PA beam and the observed PA resonances of vibrational levels in the $^1\Sigma_u^+$ state (see Fig. 1). The remarkable feature

of the PA spectroscopy of Yb atoms is that the simple structure of the spin-zero ground state 1S_0 as well as the lack of the fine and hyperfine structures for the 1P_1 excited state simplifies the comparison between the experimentally observed PA spectra and the theoretical prediction, which is in good contrast with the case of alkali atoms [3]. The $^1\Sigma_u^+$ state connects asymptotically to the $^1S_0 + ^1P_1$ state in the dissociation limit, and the long-range potential $U(R)$ at internuclear distance R is determined by the resonant dipole-dipole interaction [11]

$$U(R) = D - \frac{C_3}{R^3}, \quad C_3 = \frac{3\hbar\lambda^3}{16\tau\pi^3}, \quad (1)$$

where D is the energy in the dissociation limit, λ is the transition wavelength of the 1S_0 - 1P_1 transition, and τ is the radiative lifetime of the 1P_1 state. In addition, PA resonances ranging from near to the dissociation limit to

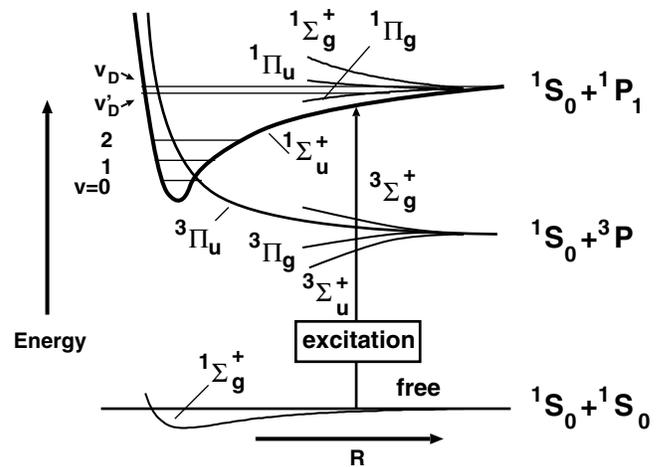


FIG. 1. Pair potential of Yb_2 . The PA light of about 399 nm excites the two free 1S_0 atoms to the molecular excited state $^1\Sigma_u^+$ which connects asymptotically to the $^1S_0 + ^1P_1$ atomic state in the dissociation limit. The long-range potential of the $^1\Sigma_u^+$ state is determined by the resonant dipole-dipole interaction $-C_3/R^3$.

the deeply bound states could be successfully observed by exploiting the high-density atoms in the FORT, rather than the atoms in a magneto-optical trap (MOT) as in the experiment of ^{40}Ca where only about 40 resonances near the dissociation limit could be observed [6]. The observed resonance frequencies were consistent with the theoretical prediction, from which we derived the atomic radiative lifetime of the $(6s6p)^1P_1$ state in much greater accuracy than previous data. Successful observation of deeply bound vibrational levels also enabled us to find the evidence of the predissociation process which was predicted as the important common feature to the alkaline-earth like atoms. Furthermore, we observed the decrease of the PA signal intensity at a large detuning from the dissociation limit. This observation, combined with the result obtained from ^{174}Yb BEC experiment, enabled us to limit the range of the scattering length.

The experimental setup is schematically shown in Fig. 2. First, a ^{174}Yb atomic beam from an oven was decelerated by the Zeeman slower using the injection-locked GaN laser diode system [12]. The Zeeman-slowed ^{174}Yb atoms were then cooled and trapped in a MOT using the intercombination transition (1S_0 - 3P_1) [13]. Typically there were 10^7 atoms in the MOT, and the peak density was about 10^{12} cm^{-3} . The temperature of the atoms was about $40\text{ }\mu\text{K}$. The atoms were transferred to the single beam FORT by a diode-pumped solid state laser whose wavelength and output power were 532 nm and 10 W , respectively [7]. The FORT beam was focused to the beam waist of $14\text{ }\mu\text{m}$, which gives the potential depth of about 1 mK . The number of the atoms trapped in the FORT was 10^6 , and the atom density was 10^{13} cm^{-3} . The temperature of the atoms was about $100\text{ }\mu\text{K}$. The decay time of the trapped atoms was about 400 ms due to the evaporation as well as the background gas collision at the vacuum of about 10^{-9} Torr . It is to be noted that the

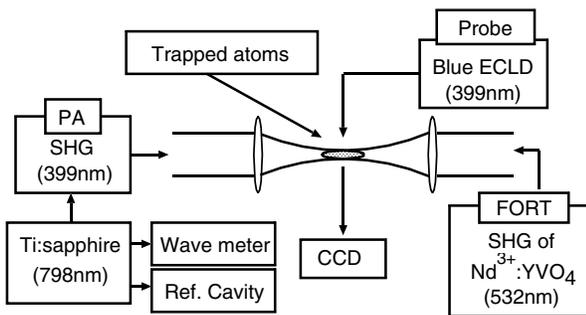


FIG. 2. Schematic experimental setup. The dense and cold Yb atoms were trapped in the FORT after the MOT with the intercombination transition. The light for the PA was generated by the frequency-doubled Ti:sapphire laser that was applied to the atoms in the FORT. The number of the atoms remaining after the irradiation of the PA beam was measured by the absorption imaging method with the charge-coupled device (CCD).

high atom density provided by the FORT was essentially important in our PA experiment.

The 399 nm light for the PA was generated by a frequency-doubled Ti:sapphire laser, the frequency of which was offset locked to an absolutely stabilized reference cavity. The laser linewidth of about 1 MHz was much smaller than the atomic linewidth of 29 MHz for the 1S_0 - 1P_1 transition. The PA beam, which propagated in the opposite direction to the FORT beam, was focused and overlapped with the FORT beam. The beam waist was $200\text{ }\mu\text{m}$ and the beam intensity was up to 100 mW . The PA beam was applied for about 200 ms during the FORT. We observed the PA signal by measuring the number of the survived atoms after applying the PA beam. The number of survived atoms were measured with the absorption imaging method using the 1S_0 - 1P_1 transition. The laser frequency was measured by a calibrated wave meter which also allowed absolute frequency measurements with an accuracy of better than 100 MHz .

We observed more than 90 PA resonances of vibrational levels in the $^1\Sigma_u^+$ state, including 80 consecutive series, up to 490 GHz detuning with respect to the atomic resonance. Figure 3 shows a typical PA signal of ^{174}Yb atoms. This resonance was assigned to $\nu'_D - \nu = 177$, where ν is the vibrational quantum number and ν'_D is the integer part of ν_D , which is an integration constant that can be interpreted as the hypothetical noninteger quantum number of the dissociation limit (see Fig. 1). The signal was fitted with the Lorentzian curve with the width of 2.9 GHz . 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 $\nu'_D - \nu = 177$ resonance

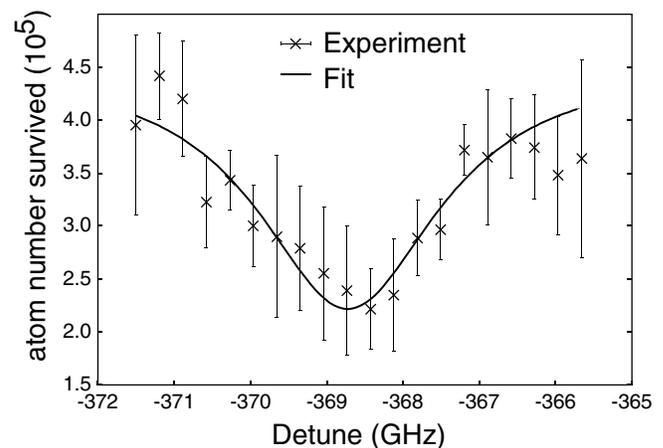


FIG. 3. PA signal of ^{174}Yb atoms. The number of the atoms in the FORT indicated by crosses was plotted against the detuning from the $^1S_0 + ^1P_1$ asymptote. Solid line is the Lorentzian fit. This resonance was assigned to the vibrational quantum number of $\nu'_D - \nu = 177$. The PA light intensity was 30 mW .

which has 3 GHz linewidth. The lack of the rotational structure also comes from the fact that the temperature of 100 μ K of the atoms trapped 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 shows the observed resonance frequencies $\Delta(v)$ relative to the $^1S_0 + ^1P_1$ asymptote as a function of the assigned numbers $v'_D - v$ in the range of 103 to 174, in which the line broadening is small enough to determine the PA frequency of the resonances. The vibrational energy structure $E(v)$ near the dissociation limit can be given by the semiclassical formula [14]

$$E(v) = D - X_0(v_D - v)^6, \quad (2)$$

$$X_0 = \frac{h^6}{\mu^3 C_3^2} \left[\frac{\Gamma(4/3)}{2\sqrt{(2\pi)}\Gamma(5/6)} \right]^6, \quad (3)$$

where μ is the reduced mass. We have obtained an excellent fit of Eq. (2) with the data where most of the deviations were less than 0.5%. The fitting curve with the function $\Delta(v) = X_0(v_D - v)^6/h$ is also shown in Fig. 4. Here we did not include the rotational energy structure due to its smallness as mentioned above. 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 2 orders of magnitude improvement in accuracy compared with the previous measurement of level crossings [15].

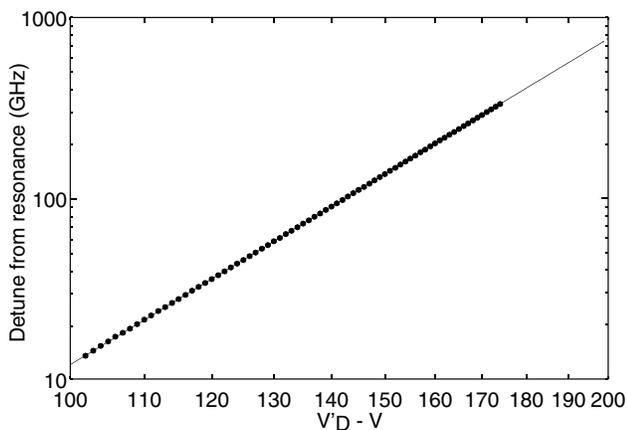


FIG. 4. Resonance frequencies of the consecutive series as a function of the assigned vibrational quantum numbers $v'_D - v$ in the range of 103 to 174, in which the line broadening is small enough to determine the PA frequency of the resonances. Both axes are in log scale. The observed detuning with respect to the $^1S_0 + ^1P_1$ asymptote indicated by closed circles was plotted against the assigned vibrational quantum number $v'_D - v$. Solid line is the fit with the semiclassical formula of Eq. (2) in the text.

Various effects such as a retardation effect and higher dispersion terms such as C_6/R^6 and C_8/R^8 would deviate the PA resonances from Eq. (2) considering only the dipole-dipole interaction term C_3/R^3 . The deviation due to these effects are estimated to be less than the residuals of the fitting of about 1 GHz (0.5% of the detunings). This fitting error mainly comes from the uncertainty of the resonance position due to the considerable line broadening of the PA resonance up to several GHz, which we discuss below. As a result, it is very difficult at present to quantitatively discuss the effects other than the C_3/R^3 term.

We have also observed line broadening of the PA 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, the resonance of $v'_D - v = 148$ had 630 MHz linewidth, and the $v'_D - v = 177$ resonance had 2.9 GHz, which is shown in Fig. 3.

We have investigated the origin of the observed line broadening. We observed almost the same linewidth of 2.2 GHz both for the PA light intensities of 8 W/cm² and 50 W/cm², which shows that the observed broadening originated from the mechanism other than the power broadening by the PA beam. Furthermore, possible unresolved rotational structures are too small to explain the observed line broadening, as was discussed above. We believe that the predissociation from the $^1\Sigma_u^+$ state to the triplet molecular states is responsible for the observed line broadening. The Yb₂ molecular potential calculated by Wang *et al.* [16] 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 $^1S_0 + ^3P$ states in the dissociation limit (see also 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 $^1S_0 + ^3D$ states, which are not shown in Fig. 1, would also be possible. The linewidth caused by the predissociation Γ can be expressed by [17]

$$\Gamma = (\Delta E_{v,v-1}/h) \times P, \quad (4)$$

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. From our measurements, the probability P was estimated to be about 0.2. This is compared with the theoretical calculation by Machholm *et al.* which predicts $P(^1\Sigma_u^+ \rightarrow ^3\Pi_u) \sim 0.64$ [18]. The agreement between the theory and the experiment on this level is impressive when we consider many approximations in the calculation. The considerably fast predissociation process Γ^{-1} compared with radiative lifetime $\tau/2$ of the $^1\Sigma_u^+$ state suggests that most of the

$^1\Sigma_u^+$ state molecules should be predissociated before radiatively decaying to the ground state $^1\Sigma_u^+$. It is, therefore, easily performed to directly detect the dissociating atoms that would be in the 3P or 3D states, as in the experiment using K atoms [17]. This would finally fix the routes of the predissociation.

We found that the amplitudes of the PA spectra monotonously decreased as the vibrational quantum number decreased. However, we have observed the irregular decrease of the intensities of PA signals only around 435 GHz detuning, which corresponds to the Condon point of 2.95 nm. The signal intensity at 435 GHz is about 20% of the normal one, for example, at 380 GHz, and the signal intensity at 465 GHz was recovered to about 80% of the one at 380 GHz. Since the intensity of the PA signal is proportional to the square of the ground-state wave function at the Condon point [19], the observed reduction of the intensity at the 435 GHz detuning indicates that the ground-state wave function has a node at 2.95 nm.

There are two probable explanations for the node of the wave function, as discussed in detail in Refs. [19,20]. The first probable explanation is related to scattering length. When the scattering length a is positive and large enough, the outmost node appears close to the value of a [19]. From this explanation, we can conclude that the scattering length is equal to 3 nm.

The other probable explanation is the oscillation of the ground-state wave function with the attractive van der Waals potential $-C_6/R^6$. This oscillation occurs at $R \leq R_Q = 1/2(mC_6/\hbar^2)^{1/4}$, where m is the mass of the ^{174}Yb atom [19]. In the above expression of R_Q , the value of C_6 of the Yb atom is not known, but can be estimated from London's formula [21] as $C_6 = 2.82 \times 10^5 \text{ MHz nm}^6$, which gives R_Q of about 4 nm. This is apparently close to the node of 2.95 nm. Since R_Q is the lower limit for the appearance of the first node, an intensity drop would also be expected already at detunings smaller than 435 GHz. Taking account also of the uncertainty of the estimated value of C_6 , however, we could not definitely conclude that the appearance of the node of the wave function is related to the wave function oscillation due to the attractive $-C_6/R^6$ potential.

Although the investigation of the signal intensity at larger detunings is quite helpful for the determination of the above two explanations, the considerable line broadenings made the observation of the signal at larger detunings quite difficult. Since we did not observe a decrease of the intensity at smaller detunings at which the Condon point is more than 3 nm, however, we can conclude that the scattering length is equal to or less than about 3 nm. In our achievement of the ^{174}Yb BEC, the scattering length of the ^{174}Yb was estimated to be 6_{-5}^{+10} nm [8]. These two pieces of information are combined to limit the ^{174}Yb scattering length in the range from 1 to 3 nm.

It is interesting to extend the PA measurements to the isotopes other than ^{174}Yb of the present work. Possible

large variation of scattering lengths among the isotopes will be a great advantage of working with the Yb atom which features a rich variety of the isotopes (five bosons and two fermions).

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