Feshbach-Resonance-Enhanced Coherent Atom-Molecule Conversion with Ultranarrow Photoassociation Resonance

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(Received 4 September 2015; published 27 January 2016)

We reveal the existence of high-density Feshbach resonances in the collision between the ground and metastable states of ^{171}Yb and coherently produce the associated Feshbach molecules by photoassociation. The extremely small transition rate is overcome by the enhanced Franck-Condon factor of the weakly bound Feshbach molecule, allowing us to observe Rabi oscillations with long decay time between an atom pair and a molecule in an optical lattice. We also perform the precision measurement of the binding energies, which characterizes the observed resonances. The ultranarrow photoassociation will be a basis for practical implementation of optical Feshbach resonances.

DOI: 10.1103/PhysRevLett.116.043202

Remarkable advances in study on ultracold atomic gases owe much to their high controllability over various experimental parameters. A tunability of interatomic interactions with Feshbach resonances (FRs) [1] has even made it possible to create ultracold molecules. By adiabatically sweeping a magnetic field across a resonance, one can create near-threshold Feshbach molecules which play a key role in many studies such as the crossover between a Bose-Einstein condensate (BEC) and a Bardeen-Cooper-Schriefer superfluid [2–4], creation [5,6], and detection [7–9] of correlated states of atoms in optical lattices. Association of Feshbach molecules is also used as the initial step to form ultracold (polar) molecules in their rovibronic ground states [10,11], which has opened the way toward ultracold chemistry [12] and quantum computation [13]. Photoassociation (PA) is another standard method to create ultracold molecules in electronically excited states with a simple optical excitation [14]. However, the transition probability is never large enough even for a strong electric-dipole-allowed ($E1$) transition due to small Franck-Condon overlap between the wave functions of free atoms and molecular bound states. In addition, the radiative lifetime of the created molecular states is usually quite short for an $E1$ transition. As a consequence, coherent production of molecules by one-color PA has been impossible until recent demonstration with a BEC of ^{88}Sr [15], where the narrow $^1S_0 \leftrightarrow {}^3P_1$ transition was exploited.

In this Letter, we report on photoassociative creation of ultracold molecules associated with the ground $^1S_0$ and metastable $^3P_2$ states of ^{171}Yb with a radiative lifetime of longer than 1 sec. Importantly, the otherwise quite small strength of the PA resonance is significantly enhanced by working around FRs for the closed channel. This is in good contrast to the technique of Feshbach-optimized PA [16–18], where a FR in an entrance channel is utilized to enhance the PA rate. The strong optical coupling enables us to demonstrate Rabi oscillations in the PA transition with a lifetime reaching hundreds of microseconds. Moreover, by using this narrow-line PA, observed FRs are characterized by precise measurement of the binding energies of near-threshold Feshbach molecules. Our observation of the strong PA line with a long expected radiative lifetime opens up the possibility to suppress atom loss in optical Feshbach resonances [19,20].

Our work is based on the existence of FRs in collisions between the $^1S_0$ and $^3P_2$ states of ^{171}Yb. Most of the currently studied FRs are for alkali atoms, which originate from their hyperfine structures with isotropic van der Waals interactions. On the other hand, the emergence of FRs associated with the existence of anisotropic interactions was first investigated in collisions of Cr [21,22], followed by the recent theoretical [23] and experimental work on Er [24] and Dy [25]. For these atoms, anisotropy in electrostatic van der Waals potentials with non-zero electronic orbital angular momenta as well as dipole-dipole interactions induce couplings to many closed channels with higher partial waves, leading to an extremely high density of FRs [26]. In the present case of the $^1S_0$-$^3P_2$ collision of Yb atoms, anisotropy originates purely from the electrostatic interaction [27].

We start with trap loss spectroscopy to locate FRs. Figure 1(a) shows the relevant energy diagram and the experimental sequence. An ultracold Fermi gas of ^{171}Yb with two spin components ($|g_\uparrow\rangle = |^1S_0(m_I = +1/2)\rangle$ and $|g_\downarrow\rangle = |^1S_0(m_I = -1/2)\rangle$) is prepared by sympathetic evaporative cooling with ^{173}Yb in an optical dipole trap with a wavelength of 532 nm [28]. After evaporation we obtain a thermal gas of $8.3(2) \times 10^4$ atoms at a temperature $T = 1.1(1)$ μK. The trap frequencies are $(\omega_x, \omega_y, \omega_z)/2\pi = (327, 62.4, 426)$ Hz with z indicating the direction of gravity, leading to the Fermi temperature of $T_F = 620$ nK. Remaining ^{173}Yb atoms are removed by 556 nm light at the resonant frequency of the $^1S_0 \rightarrow {}^3P_1(F = 7/2)$ transition. At a static magnetic field of $B = B_z = 1.0$ G, we excite a

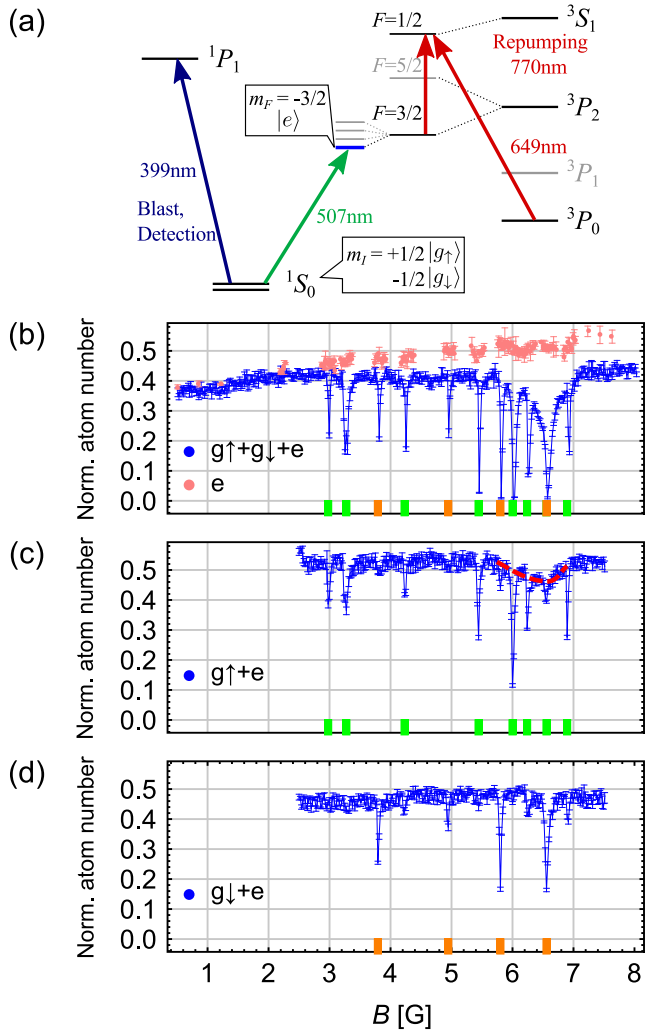

FIG. 1. (a) Low-lying energy level diagram of ^{171}Yb. (b)–(d) Observation of FRs (marked with rectangles) in a mixture of $|g_\uparrow\rangle$-$|g_\downarrow\rangle$-$|e\rangle$, $|g_\uparrow\rangle$-$|e\rangle$ and $|g_\downarrow\rangle$-$|e\rangle$, respectively. Trap loss spectra at temperature of (b) 1.1 and (b),(c) 1.8 μK are shown. Pink points in (b) represents the data for a sample containing $|e\rangle$ atoms only. Red dashed line in (c) highlights the existence of a broad resonance at 6.6 G.

small fraction of atoms to the lowest hyperfine state in $^3P_2$, $|e\rangle = |^3P_2(F = 3/2, m_F = -3/2)\rangle$ state, by applying resonant laser light at 507 nm [29]. Immediately after the excitation, we ramp up the field to the desired value within 10 ms. After a hold time of 100 ms in the dipole trap, atoms in the ground state are removed, followed by repumping of survived atoms in the $^3P_2$ state into the ground state. The repumped atoms are detected by fluorescence imaging with a magneto-optical trap using 399 nm cooling light.

Figure 1(b) shows 12 resonant losses which are attributed to the existence of FRs in $|g_\uparrow\rangle$-$|e\rangle$ or $|g_\downarrow\rangle$-$|e\rangle$ collisions [30]. We exclude the possibility of $|e\rangle$-$|e\rangle$ resonances by repeating the same experiment with $|e\rangle$ atoms only. Loss spectra for the $|g_\uparrow\rangle$-$|e\rangle$ and $|g_\downarrow\rangle$-$|e\rangle$ states are also shown in Figs. 1(c) and 1(d), respectively, by which we can identify the relevant spin combination for an open channel in each resonance. For spin polarization, we apply optical pumping with the $^1S_0 \rightarrow {}^1P_1$ $(F = 1/2)$ transition at the early stage of evaporative cooling. Because of the incompleteness of optical pumping, resonances with the residual spin component are also visible for each case of two-component spectrum. Optical Stern-Gerlach experiment infers the spin distribution to be $(g_\uparrow, g_\downarrow, e) = (30\%, 50\%, 20\%)$, (70%,10%,20%), and (5%,75%,20%) for Figs. 1(b), 1(c), and 1(d), respectively, which accounts for the contamination of weak resonances. We note that the strong loss feature at 6.6 G seen in Fig. 1(b) is the consequence of two overlapping resonances: the broad resonance in the $|g_\uparrow\rangle$-$|e\rangle$ collision and the narrow resonance in the $|g_\downarrow\rangle$-$|e\rangle$ collision. As shown in Fig. 1(c), the former FR is accompanied by relatively small loss compared to the other narrow ones, which possibly reflects the open-channel dominated feature of this resonance. The broad resonance with small loss is favorable for practical use. Exploring FRs for fermionic isotopes is of great importance because the stability of resonantly interacting two-component Fermi gases provides much wider practical applications. Especially in the case of $^1S_0$-$^3P_2$ resonances, using polarized fermions is the best way to avoid inelastic collisions in the $^3P_2$ state which is a dominating decay channel [34].

By working around the observed FRs, we directly form ultracold molecules with a PA method. Owing to the ultranarrow $^1S_0 \leftrightarrow {}^3P_2$ line in our PA method, it is possible to resolve molecular resonance down to ~10 kHz, which is Doppler-limited linewidth of our samples. Figure 2(a) shows a $|g_\downarrow\rangle \rightarrow |e\rangle$ laser spectrum taken at $B = 6.2$ G, in the vicinity of broad $|g_\uparrow\rangle$-$|e\rangle$ resonance at 6.6 G. In this measurement, we simply irradiate excitation laser onto a ground state $|g_\uparrow\rangle$-$|g_\downarrow\rangle$ mixture at $T = 65(6)$ nK $(T/T_F = 0.27)$ and measure the number of atoms remaining in the ground state by absorption imaging. On the red-detuned side of the atomic resonance, we clearly observe a resonant loss of atoms. This resonance disappears when spin-polarized samples are used, which indicates photo-induced association of molecules from colliding $|g_\uparrow\rangle$-$|g_\downarrow\rangle$ pairs. Large spatial extent of the Feshbach molecule provides the large Franck-Condon factor, giving PA rate comparable to the atomic transition.

The frequency detuning of the PA resonance from the atomic resonance directly gives the binding energy of the associated molecules. We systematically measure the magnetic field dependence of the binding energy and confirm that the observed molecular state is associated with the FR at 6.6 G, as shown in Fig. 2(b). In addition, we find that another molecular level stems from the resonance at 6.0 G and its anti level crossing with one from the 6.6 G resonance [Fig. 2(c)]. For further characterization of the observed FR, we perform coupled two-channel calculation [35] of the binding energy. In the calculation, we fix the

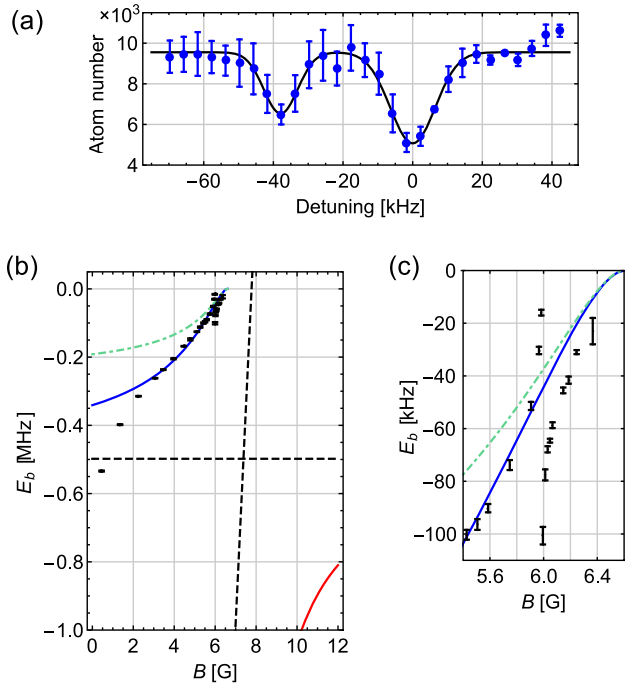

FIG. 2. (a) $^1S_0 \leftrightarrow {}^3P_2$ laser spectroscopy at $B = 6.2$ G. The PA resonance to the Feshbach molecule associated with the 6.6 G broadest FR is visible at -38 kHz from the atomic resonance. The PA laser pulse has a duration 20 ms and intensity 2.3 W/cm^2. (b) Binding energy of the Feshbach molecule associated with resonances at $B = 6.0$ and 6.6 G. The solid lines are the result of the coupled two-channel calculation. Two dashed lines indicate the bare closed channel molecule and the shallowest bound state of the open channel. The dot-dashed line is the prediction of the universal relation $E_b = -\hbar^2/(ma^2)$. (c) Detailed behavior near the anti level crossing at 6 G.

unknown parameters $C_6 = 3000$ a.u. for the $^1S_0$-$^3P_2$ interatomic potential and the differential magnetic moment between the open and the closed channel $\delta\mu = 0.9\mu_B$, where μ_B is the Bohr magneton. We find that the calculation with background scattering length $a_{\rm bg} = 290a_0$ and the resonant width $\Delta = 1.11$ G well reproduce the experimentally determined binding energy. In the low field $B < 3$ G, the data shows deviation from the theory, which indicates the existence of the coupling to unknown molecular levels. As seen in the figure, the binding energy of the Feshbach molecule remarkably deviates from that of the bare closed-channel molecule. Therefore, the binding energy is not sensitive to the magnitude of $\delta\mu$, which prevents us from extracting the magnetic moment of the closed channel.

To directly observe the induced change of the scattering length, we perform laser spectroscopy on the $^1S_0 \leftrightarrow {}^3P_2$ transition also in an optical lattice. In a deep optical lattice where each lattice site can be regarded as an isolated potential well, resonance from doubly occupied sites is shifted by the on-site interaction $U_{g,e} - U_{g,g}$ which can be converted to the scattering length $a_{g,e}$ [27,36]. We adiabatically load a degenerate spin mixture of $|g_\uparrow\rangle$ and $|g_\downarrow\rangle$ at $T/T_F = 0.17(2)$ into a simple cubic optical lattice with a wavelength $\lambda = 532$ nm. After reaching the lattice depth of 14.7 times the recoil energy $E_R = \hbar^2(2\pi/\lambda)^2/(2m)$, we apply an excitation pulse with intensity 0.12 W/cm^2 and duration 1 ms to excite $|g_\downarrow\rangle$ atoms, followed by the repumping scheme to count $|e\rangle$ atoms. Because of the Pauli principle, only the combination of $|g_\uparrow\rangle$-$|e\rangle$ is produced from doubly occupied sites and the corresponding scattering length $a_{g_\uparrow,e}$ can be measured.

Figure 3(a) is a typical spectrum in the lattice, which shows three prominent peaks. The largest peak corresponds to the excitation of isolated atoms, accompanied by the blue sideband resonance separated by the band gap ~29 kHz. One more resonance found between these two resonances is the signal from the doubly occupied sites. We determine the scattering length $a_{g_\uparrow,e}$ by using the analytic model of a harmonically trapped atom pair [37] as

$$a_{g_\uparrow,e} = \frac{a_{\rm ho}}{\sqrt{2}} \frac{\Gamma(-U_{g_\uparrow,e}/(2\hbar\omega) - 1/2)}{\Gamma(-U_{g_\uparrow,e}/(2\hbar\omega))}, \qquad (1)$$

where the on-site interaction $U_{g_\uparrow,e}$ is given by the $U_{g_\uparrow,e} = 2\pi\hbar\Delta f + U_{g_\uparrow,g_\downarrow}$ with Δf denoting the frequency difference between the singly and the doubly occupied sites and $U_{g_\uparrow,g_\downarrow} = -2\pi\hbar \times 82$ Hz the on-site interaction in the ground state obtained by the known scattering length $a_{g_\uparrow,g_\downarrow} = -0.15$ nm [38]. The mean trap frequency $\omega/2\pi = 28.3$ kHz of each potential well is deduced from the band calculation [30].

We map out the magnetic field dependence of $a_{g_\uparrow,e}$ around the 6.6 G FR, as shown in Fig. 3(b). In addition to the resonant change of the scattering length at 6.6 G, the effect of the narrower 6.0 G resonance is also visible. We fit the obtained field dependence with the model proposed in Ref. [39],

$$a(B) = a_{\rm bg}\left(1 - \frac{\Delta_2}{B - B_2} - \frac{\alpha\Delta_1}{B - B_1}\right), \qquad (2)$$

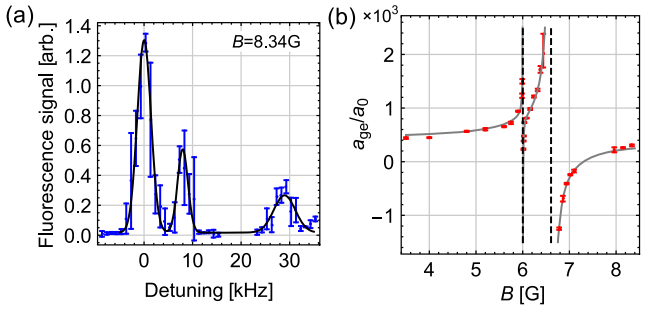

FIG. 3. (a) Spectrum on the $^1S_0 \leftrightarrow {}^3P_2$ transition in an optical lattice with the depth of $14.7E_R$. (b) Magnetic field dependence of the scattering length $a_{g\uparrow,e}$ determined by spectroscopy in an optical lattice.

where $\alpha = [(B_1^{(0)} - B_2^{(0)})/(B_1^{(0)} - B_2^{(0)} - \delta B_2)]^2$. Here $B_1^{(0)}$ ($\simeq B_1$) and $B_2^{(0)}$ are the zero-crossing points of bare molecular energies. For the shift $\delta B_2 = B_2 - B_2^{(0)}$, we adopt an approximation [35]

$$\delta B_2 \simeq \Delta_2 \frac{(a_{\rm bg}/\bar{a})(1 - a_{\rm bg}/\bar{a})}{1 + (1 - a_{\rm bg}/\bar{a})^2}, \tag{3}$$

with the mean scattering length $\bar{a} = 4.4$ nm evaluated from the C_6 constant mentioned above. Equation (2) is valid when the width of the first resonance is much narrower than that of the second resonance, $|\Delta_1| \ll |\Delta_2|$. The fit yields $B_1 = 6.003(5)$ G, $\Delta_1 = 3.7(3)$ mG, $B_2 = 6.607(9)$ G, $\Delta_2 = 680(40)$ mG, and $a_{\rm bg} = 406(15)a_0$. Obtained parameters slightly differ from that inferred from the binding energy measurement. Possible source of the uncertainty in the scattering parameters is the existence of unknown resonances at higher fields [30]. Especially, extracting the value of $a_{\rm bg}$ from the fit is highly sensitive to tails of unknown resonances.

Narrow linewidth of the PA transition and resulting long lifetime of an associated molecule enable coherent production of molecules. For the narrow line ($\Gamma/2\pi = 7$ kHz) PA of a ^{88}Sr BEC, coherent Rabi oscillations up to $\sim$10 μs were reported [15]. Isolated atom pairs residing on each site of an optical lattice are more suitable system to observe coherent phenomena, for unwanted inelastic collisions are suppressed and long lifetime of molecules can be achieved [40].

We demonstrate the Rabi oscillations between $|g_\uparrow\rangle$-$|g_\downarrow\rangle$ atom pairs and $|g_\uparrow\rangle$-$|e\rangle$ Feshbach molecules via one-color PA. Figure 4(a) shows both the atomic and atom-molecule Rabi oscillations in the optical lattice with $14.7E_{\rm R}$ depth.

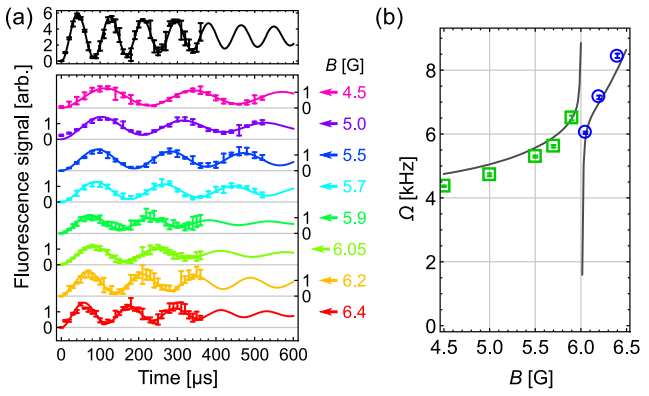

FIG. 4. (a) Rabi oscillations of isolated atoms (top) and atom pairs $\leftrightarrow$ molecules at various magnetic field (bottom) in an optical lattice. The laser intensity is fixed to 58 W/cm^2. For the atomic transition, the Rabi frequency is 11.8 kHz. (b) The atom-molecule Rabi frequency Ω as a function of the magnetic field. Symbols (circles, squares) distinguish the different branches of the anticrossing at 6 G. The solid line shows the prediction from the wave function overlap (see text).

Since the $|g_\downarrow\rangle \rightarrow |e\rangle$ transition is $E1$ allowed, the Rabi frequency is $\sim$10 times larger than the purely $M2$-allowed case [31], which makes it much easier to observe clear oscillations. The e^{-1} decay time of the oscillations is on the order of 500 μs, which is limited by the inhomogeneous broadening of the resonant frequency. The observed atom-molecule Rabi frequency is on the same order as that of the atomic transition and retains reasonable magnitude over the range of several gauss. To account for the field dependence of the Rabi frequency, we carry out the following simple analysis. The optical coupling between the initial and final states is proportional to $\langle g_\uparrow, e|d(R)|g_\uparrow, g_\downarrow\rangle$, where $d(R)$ is the transition dipole depending on the interatomic distance R [41]. For a weakly bound Feshbach molecule, $d(R)$ is almost unchanged from its asymptotic value, i.e., the atomic transition dipole, over the most range of molecular extent. Therefore, the Rabi frequency can be reduced to the simple product $\Omega = \Omega_{\rm atom} \times |\langle g_\uparrow, e|g_\uparrow, g_\downarrow\rangle|$. Using the analytic expression for the two-particle wave functions in a harmonic potential [37] and the experimentally determined scattering length, we find good agreement between the data and the above estimation [Fig. 4(b)].

In conclusion, we observed the FRs between the ground and metastable excited states of ^{171}Yb and successfully formed associated Feshbach molecules by one-color PA. Observed density of resonances, 0.7/G per spin combination, is lower than the cases of highly magnetic Er and Dy, while much higher than the typical of alkali-metal atoms. This may be the consequence of the simpler level structure and less anisotropic interactions of ^{171}Yb. Moderately dense resonances are convenient for tuning interactions with a selected well-behaved resonance and for future theoretical analysis of the observed resonances. The Feshbach molecule can be strongly coupled to the ground-state scattering wave function via the PA transition. Optical Feshbach resonance on this transition will effectively change the interaction between the ground state with a small atomic loss. Finally, we note that ultranarrow PA demonstrated here is applicable not only to the homonuclear collisions of alkaline-earth-metal-like atoms, but also to the heteronuclear collisions between alkali atoms and alkaline-earth-like atoms, such as Li-Yb [42].

This work was supported by the Grant-in-Aid for Scientific Research of JSPS (No. 25220711, No. 26247064) and the Impulsing Paradigm Change through Disruptive Technologies (ImPACT) program by the Cabinet Office, Government of Japan.

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