Controlled Production of Subradiant States of a Diatomic Molecule in an Optical Lattice

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We report the successful production of subradiant states of a two-atom system in a three-dimensional optical lattice starting from doubly occupied sites in a Mott insulator phase of a quantum gas of atomic ytterbium. We can selectively produce either a subradiant 1_g state or a superradiant 0_u state by choosing the excitation laser frequency. The inherent weak excitation rate for the subradiant 1_g state is overcome by the increased atomic density due to the tight confinement in a three-dimensional optical lattice. Our experimental measurements of binding energies, linewidth, and Zeeman shift confirm the observation of subradiant levels of the 1_g state of the Yb_2 molecule.

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Spontaneous emission of dipole radiation from two identical atoms is a fundamental process theoretically studied by R. H. Dicke, who introduced the important concepts of superradiance and also subradiance [1]. While both of these correspond to quantum superpositions of product states with a ground state in one atom and an excited state in another, the spontaneous emission is enhanced (suppressed) in the superradiant (subradiant) state due to constructive (destructive) interference. Although the phenomenon of superradiance is widely observed in various physical systems, subradiance is not well studied [2]. The difficulty of creating and studying the subradiant state naturally comes from its reduced radiative interaction.

In this Letter, we report our successful controlled production of a subradiant state of a two-atom system in single quantum states of both internal ro-vibronic and external center-of-mass degrees of freedom, starting from doubly occupied sites in a Mott insulator phase of an atomic quantum gas in a three-dimensional optical lattice. For the case we study, the subradiant and superradiant states correspond, respectively, to 1_g and 0_u states, labeled by the standard molecular Hund's case (c) notation. By using a two-electron atom with no structure in the ground state, we realize an ideal situation for observing and studying a subradiant 1_g state. In addition, the inherent weak excitation rate is overcome by the tight confinement of two atoms in a three-dimensional optical lattice. A suppressed spontaneous emission rate is confirmed by the linewidth measurement as one of the important evidences of the subradiance. We can selectively produce either a subradiant 1_g or superradiant 0_u state by choosing the excitation laser frequency. In addition, observed binding energies are in excellent agreement with theoretical analyses. While it might be possible to populate these subradiant molecular states by some simpler scheme in a uncontrolled or nonselective way, it should be difficult to study the system with high precision and controllability. We believe that our highly controlled production scheme will open the door to a number of interesting experiments. For example, the inherent sub-kHz spectrum linewidth of the optical transition from the ground state to these subradiant molecular states is straightforwardly applicable to a high-resolution photoassociation spectroscopic study of a quantum many body state in an optical lattice. In addition, our scheme will offer a novel possibility of creating a long-lived electronically excited molecular quantum gas in which the collisional stability can be studied.

The studied subradiant state of the two-atom system corresponds to a diatomic molecular state with an inversion symmetry, which is one of the fundamental symmetries exhibited by a homonuclear diatomic molecule. Quantum states whose wave function retains its sign upon inversion of electron coordinates are labeled *gerade*, as opposed to *ungerade* states, whose wave function changes sign after inversion. We write two degenerate molecular excited states: $|A(P)B(S)\rangle$ and $|A(S)B(P)\rangle$, where S and P represent the ground and excited states, respectively, and A and B represents two atoms. As molecular eigenstates we consider the symmetrized molecular states,

$$|\Psi_\pm\rangle = \frac{1}{\sqrt{2}}[|A(P)B(S)\rangle \pm |A(S)B(P)\rangle], \tag{1}$$

where, the $|\Psi_+\rangle$ state is of ungerade symmetry and the $|\Psi_-\rangle$ state is gerade because the atomic excited state considered here itself is ungerade. The fact that the dipole operator, whose matrix elements determine the transition width, is itself ungerade leads to the Laporte selection rule [3] which states that optical dipole transitions are only possible between states of opposite inversion symmetry. The *gerade-gerade* (g-g) transition is, therefore, forbidden, whereas a *gerade-ungerade* (g-u) one is allowed, having a natural linewidth twice the bare atomic one Γ_{at}. These are nothing but the diatomic molecular realizations of phenomena of subradiance and superradiance.

It is noted that a retardation effect caused by the finite speed of interaction propagation, although weak, can lead to the breakdown of the Laporte rule. An example from classic spectroscopy is the well-known Lyman-Birge-Hopfield band of the nitrogen molecule [4,5], which is the first confirmed molecular gerade-gerade transition [6]. Consider a diatomic molecule immersed in an electromagnetic field of a laser beam. If the two atoms are confined closely the electric field "felt" by the two is approximately the same. In this case, we expect considerable suppression of the spontaneous emission or optical excitation. In other words, it is very difficult to create a subradiant state by optical excitation from the ground state. In reality, however, the two atoms in molecular states close to the dissociation are weakly coupled and relatively far apart. Therefore, there is a phase difference in the electric fields experienced by them. When calculating the transition rate to the excited state from the ground state of $|A(S)B(S)\rangle$, we have to include this phase difference [7] and replace the usual transition matrix element by

$$|\langle A(S)B(S)|\hat{d}_A + \hat{d}_B \exp(-i\vec{k}\cdot\vec{R})|\Psi_\pm\rangle|^2, \qquad (2)$$

where $\vec{R}$ is the distance between the atoms and $\vec{k}$ is the incident photon's wave vector. When we consider the spontaneous emission, this has to be averaged over all possible photonic states. The transition widths γ depend on the projection Λ of the orbital angular momentum on the internuclear axis (Σ states have $\Lambda = 0$, while Π states have $|\Lambda| = 1$) and are [7–11]

$$\gamma(\Sigma) = \Gamma_{at}\left(1 \pm \frac{3}{v^3}[-v\cos v + \sin v]\right), \qquad (3)$$

$$\gamma(\Pi) = \Gamma_{at}\left(1 \pm \frac{3}{2v^3}[v\cos v - (1 - v^2)\sin v]\right), \qquad (4)$$

where $v = kR$ and "+" is for allowed g-u transitions, while "−" applies to the forbidden g-g and u-u transitions. These expressions for $\Lambda = 0$ and $|\Lambda| = 1$ states translate directly to those for $\Omega = 0$ and $|\Omega| = 1$ states in Hund's case (c) [12,13], which is relevant to the molecular states in this work, where Ω is the projection of the total electronic angular momentum j on the molecular axis. Note that in the limit of small internuclear distances ($v \rightsquigarrow 0$) these expressions lead to $2\Gamma_{at}$ for the allowed g-u transitions and zero for the forbidden g-g and u-u transitions. At infinite separations, $v \rightsquigarrow \infty$, these widths become Γ_{at}, exactly as we would expect for uncorrelated atoms.

Although Refs. [14,15] study deeply bound levels of alkali dimer, the alkali species are complicated by g-u mixing in the ground state induced by nuclear spin. Having a nondegenerate ground state is an important prerequisite for the clear demonstration of subradiance. This is satisfied in our work by using a two-electron atom of ytterbium (^{174}Yb) where the ground state is $^1S_0$.

Since the molecular levels we study have binding energies much smaller than the fine structure splitting in the excited states, they are well described by Hund's coupling case (c). The excited molecular states that are asymptotically connected to the $^1S_0 + {}^3P_1$ separated atom limit are the doubly degenerate 1_g and $1_u(|\Omega| = 1)$ states and the 0_g and $0_u(\Omega = 0)$ nondegenerate states (see Fig. 1). The subscript $g(u)$ denotes gerade (ungerade) symmetry. The 0_u and 1_g states have long-range attractive molecular potentials that support respective superradiant and subradiant bound states. We present the interaction potential V^{1_g} of two atoms in the 1_g subradiant state as [12,13,16]

$$V(R) = -\frac{C_3^{1_g} t(R)}{R^3} - \left(1 - \frac{\sigma_{1_g}^6}{R^6}\right)\frac{C_6^{1_g}}{R^6} - \frac{C_8^{1_g}}{R^8} + \frac{\hbar^2\{J(J+1)\}}{2\mu R^2}, \qquad (5)$$

where $\hbar$ is the Planck constant divided by 2π, μ is the reduced mass of the Yb atom pairs, $t(R) = \cos v + v\sin v - v^2\cos v$, and J is the quantum number for the total angular momentum. The resonant dipole-dipole

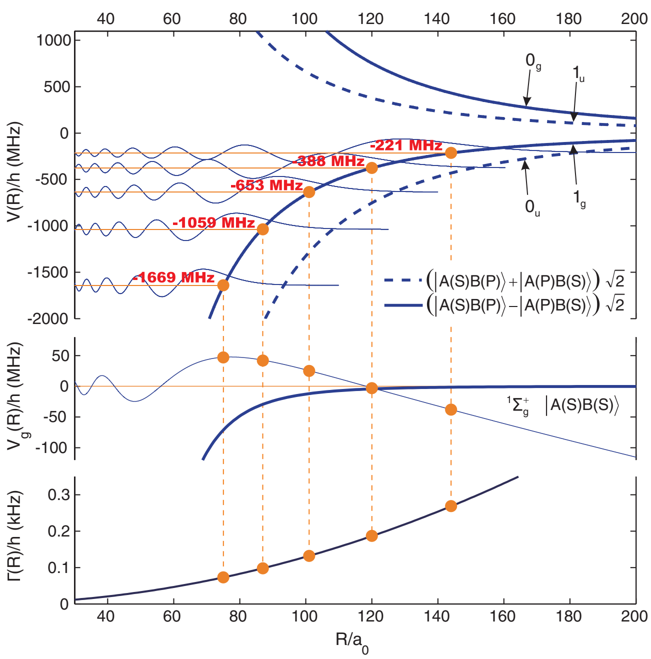

FIG. 1 (color online). Bound states in the excited 1_g state in $J = 1$. Top panel: Energies, outer turning points, and wave functions of the bound states in the investigated range. Middle panel: Ground state wave function at a collision energy of $\varepsilon_r/k_B = 1\ \mu$K. Note that the outer turning point of the −388 MHz state is almost exactly on the node of the ground state wave function, making it unobservable. Bottom panel: Natural linewidth $\Gamma(1_g)$ of the g-g transition expected in retardation theory [see Eq. (4)]. Note that for most transitions this width is less than 200 Hz.

interaction coefficient $C_3^{1_g}$ is most conveniently described in terms of the atomic transition linewidth: $C_3^{1_g} = (3/4)\Gamma_{\rm at}(\lambda/2\pi)^3$, where λ is the wavelength of the $^1S_0 - {}^3P_1$ transition. From a simple theoretical argument, the value of $C_3^{1_g}$ is one half of $C_3^{0_u}$ which is the resonant dipole-dipole interaction coefficient for the 0_u state. The terms including $C_6^{1_g}$ and $C_8^{1_g}$ represent the van der Waals interaction. The $\sigma_{1_g}^6$ parameter characterizes the strength of the short-range interaction [16].

The experimental procedure is as follows. First, we generate a ^{174}Yb Bose-Einstein Condensate [17]. Then we adiabatically ramp up a three-dimensional optical lattice potential using three mutually orthogonal laser beams with the wavelength of 532 nm [18]. The typical potential depth of the optical lattice is about $22E_R$, in which $E_R = 0.2\ \mu$K is the recoil energy for the Yb atom. At this lattice depth, the system forms a Mott insulator where the atoms are well localized at each lattice site up to the filling of three atoms per site, depending on the total atom number. Then, the linearly polarized photoassociation light which is red-detuned from the $^1S_0 - {}^3P_1$ transition of Yb atoms is focused on the atoms in the optical lattice with a beam waist of 70 μm. The typical pulse has a duration of 100 ms and a power of 200 μW. The photoassociation light induces the trap loss of the atoms when it is resonant with a molecular bound state in the excited state. The number of the atoms remaining in the trap is measured with an absorption imaging technique. The probe light has a duration of $\sim$100 μs and a power of $\sim$100 μW. The residual magnetic field is reduced to less than 0.5 G.

With this procedure, we found eight new resonances within the range of 2 GHz red-detuning, in addition to the already observed photoassociation resonances associated with the 0_u state. Figure 2 shows spectra of the newly found photoassociation resonances. The new resonances are quite different from those of 0_u and are successfully assigned as levels of the 1_g states (see Table I), as explained in detail below. The measured binding energies in Table I are compensated for the light shifts due to the photoassociation light by measuring the intensity dependence of the binding energies and extrapolating to zero intensity.

It should be also noted that we could not observe the 1_g resonances in a weak harmonic trap. When we confine atoms in the vibrational ground state of an optical lattice, however, the effective local density is increased by the ratio of one over the volume of the ground state of the optical lattice to the number of atoms over the whole trap volume of the free gas. In addition, by using the quantum gas, namely, two atoms in the lowest vibrational state in an optical lattice, the observed spectra do not suffer from the broadening due to the k_BT spread in the case of a free thermal gas with a temperature T, where k_B is the Boltzmann constant. These effects result in the conversion of a weak free-bound transition into a rather stronger bound-bound transition [19,20].

We carefully confirm that the new resonances originate from the subradiant 1_g state. The first important evidence is the spectrum linewidth. As shown in Figs. 2(b)–2(i), the

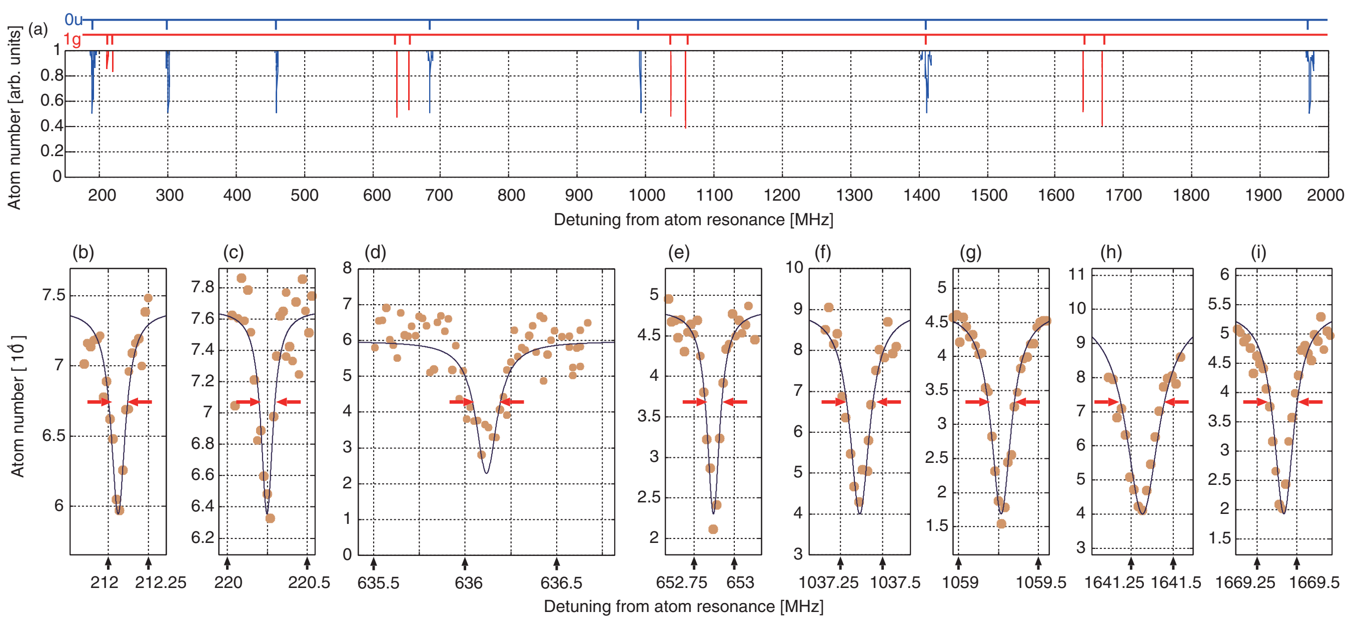

FIG. 2 (color online). (a): All photoassociation spectra of the 1_g states we observed. The 0_u states are also shown and are taken from Ref. [21]. The horizontal and vertical axes are the frequency difference of the excitation laser from the atom resonance $^1S_0 - {}^3P_1$ and the number of remaining atoms, respectively. (b)–(i): Spectra of the 1_g states. The solid line is a fit of the Lorentz function. Arrows show full widths at half maximum of the Lorentz function and the widths are (b) 0.108(17) MHz, (c) 0.088(24) MHz, (d) 0.141 (25) MHz, (e) 0.095(8) MHz, (f) 0.142(22) MHz, (g) 0.150(8) MHz, (h) 0.236(41) MHz, (i) 0.151(15) MHz, respectively.

TABLE I. Measured and calculated binding energies (E_b) for subradiant molecular states, where v and J are the vibrational and rotational quantum numbers of the states. $v' = v'_D - v$ is numbered from the dissociation limit. Values of calculated Condon points (R_C) are also shown, where a_0 is the Bohr radius.

	$J = 1$			$J = 2$		
	Exp.	Theory		Exp.	Theory	
v'	E_b (MHz)	E_b (MHz)	R_C (a_0)	E_b (MHz)	E_b (MHz)	R_C (a_0)
9	−221.23(3)	−221.13	142	−212.08(2)	−212.26	143
10	not found	−388.38	118	not found	−376.05	119
11	−652.91(3)	−652.92	100	−636.33(18)	−636.29	100
12	−1059.33(7)	−1059.36	85.3	−1037.53(7)	−1037.48	85.6
13	−1669.45(8)	−1669.51	73.9	−1641.41(16)	−1641.37	74.0

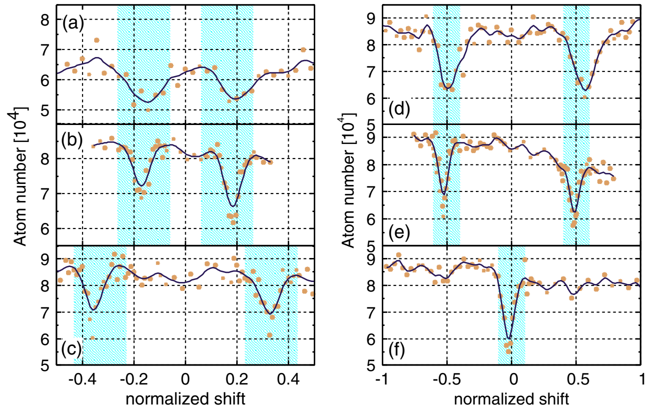

FIG. 3 (color online). Zeeman spectra of $v' = 9$, $J = 2$ resonance [(a), (b), (c)] and of $v' = 9$, $J = 1$ resonance [(d), (e), (f)] under a magnetic field applied along the x axis [(a) and (d)], y axis [(b) and (e)], and z axis [(c) and (f)]. The frequency shifts shown here are normalized by the Zeeman shift of atoms $g_{Ja}\mu_B B$. Shadow areas indicate the expected theoretical values of $m_j/[J(J+1)]$ [see Eq. (6) in the text], which are 1/6 for (a), (b), 1/3 for (c), 1/2 for (d), (e), and 0 for (f), respectively.

widths of the new resonances are narrower than the theoretical value of 2×182 kHz of the superradiant 0_u state [21], and especially the linewidths in Figs. 2(c) and 2(e) are less than 100 kHz, which are even narrower than the atomic linewidth of 182 kHz. From this feature we can exclude the possibility that the new resonance originates from single atoms or the superradiant 0_u state. Since the linewidth of our photoassociation laser is measured to be almost the same as the measured photoassociation spectrum width, the intrinsic width of the new resonance could be much narrower, consistent with the theoretically expected value of 0.2 kHz for the subradiant 1_g state (see Fig. 1).

Next, we confirm that the observed binding energies fit well with the eigenvalues of the potentials in Eq. (5) as shown in Table I. They consist of two series of $J = 1$ and $J = 2$. No resonances are assigned to $J = 0$ and $J > 2$. The fitting parameters are $C_3^{1_g} = 0.5 \times 0.193\,706 E_h a_0^3$, $C_6^{1_g} = 2283.6 E_h a_0^6$, $C_8^{1_g} = 748\,788 E_h a_0^8$, $\sigma_{1_g} = 8.6006 a_0^6$, where a_0 is Bohr radius and E_h is Hartree energy. The value of $C_3^{1_g}$, which is expected as one half of $C_3^{0_u}$, is in good agreement with $C_3^{0_u} = 0.194\,886 E_h a_0^3$ [16], which supports that the new resonances originate from the subradiant 1_g state. In our experiment, no resonances with the Condon points around $120a_0$ are observed, indicated as "not found" in Table I, since the Condon point is quite close to the node of the ground state wave function and therefore the transition rate is suppressed [22,23]. The interatomic potential and the bound state energies of the measured states are shown in the upper panel of Fig. 1. The Condon points for the transitions to these states range from $75a_0$ to $144a_0$. According to the theory, these correspond to natural linewidths Γ ranging from 100 to 250 Hz and thus are extremely weak.

Third, we investigate the Zeeman shift of the new resonance. In Hund's case (c), the Zeeman splitting energy E_Z by an external magnetic field B is calculated within first-order perturbation theory [24] as

$$E_Z = g_{\rm mol}\mu_B m_J B = \frac{\Omega^2}{J(J+1)} g_{Ja}\mu_B m_J B, \qquad (6)$$

where $g_{Ja} \sim 1.5$ is Lande's g factor of Yb atoms in the $^3P_1$ states, and μ_B is the Bohr magnetic moment. Here $|\Omega| = 1$ for the 1_g state, and therefore the 1_g states have $g_{\rm mol} = g_{Ja}/2$ for $J = 1$ and $g_{\rm mol} = g_{Ja}/6$ for $J = 2$. We experimentally measure the Zeeman shifts under an external magnetic field. The spectra around −212.08 MHz and −221.229 MHz under different magnetic field directions are shown in Fig. 3. We measure three kinds of spectra in order to study the Zeeman shifts as well as the selection rules of the transition. An external magnetic field is applied along the x, y, or z axis, where x is the axis of polarization of photoassociation light, and z is the propagation axis of the photoassociation light. The observed Zeeman splitting is consistent with our expectations: the $J = 2$ resonance is an $E2$ transition and $\Delta m = \pm 1$ is allowed for the external magnetic field along the x and the y axes, $\Delta m = \pm 2$ for the z axis. In contrast, the $J = 1$ resonance is an $M1$ transition and $\Delta m = \pm 1$ for the x and the y axes, $\Delta m = 0$ for the z axis.

In summary, we report our successful observation of optical excitation into subradiant states of the $^{174}Yb_2$ molecule. The inherent weak excitation rate is overcome by tight confinement in a three-dimensional optical lattice, which gains a bound-bound transition rate instead of free-bound. The experimental measurements of binding energies, linewidth, and Zeeman shift confirm that the origin of the newly found resonances are the subradiant 1_g state.

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