

Prospects for Making Polar Molecules with Microwave Fields

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We propose a mechanism to produce ultracold polar molecules with microwave fields. It converts trapped ultracold atoms into vibrationally excited molecules by a single microwave transition and entirely depends on the existence of a permanent dipole moment in the molecules. As opposed to production of molecules by photoassociation or magnetic-field Feshbach resonances, our method does not rely on properties of excited states or existence of Feshbach resonances. We determine conditions for optimal creation of polar molecules in vibrationally excited states of the ground-state potential by changing frequency and intensity of the microwave field. We also explore the possibility to produce vibrationally cold molecules by combining the microwave field with an optical Raman transition or by applying a microwave field to Feshbach molecules. The production mechanism is illustrated for KRb and RbCs.

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Quantum gases of polar molecules have attracted the attention of researchers due to their exceptional properties that allow them to interact over large distances by the dipole-dipole force. These properties might be observable when polar molecules are produced at ultracold temperatures. To date the coldest polar molecules (with a temperature below $1 \mu\text{K}$) have been created by either magnetic-field Feshbach resonances [1–4], photoassociation [5–11] or, most recently, radio-frequency magnetic-dipole transitions [12]. In the majority of cases these molecules are formed in highly excited vibrational levels of the electronic ground state. However, proposals that use polar molecules, either in simulation of many-body systems [13–15] or as qubits in a quantum processor [16], require these molecules to be vibrationally cold. This was first achieved in the photoassociation experiment by DeMille's group at Yale University [17]. They formed polar RbCs molecules in the $v = 0$ level of the $X^1\Sigma^+$ ground electronic potential, although the number of vibrationally cold molecules was relatively small and probably insufficient for the above mentioned applications.

The latest development in photoassociation, which promises to greatly increase the molecular production rate, is the use of an optical lattice. Experiment [18] has demonstrated that a Mott insulator phase can be formed with exactly two atoms per optical lattice site. Two such atoms can then be associated into a single molecule. The advantage of using an optical lattice is that a pair of atoms in a site is well isolated from distractive perturbations by the mean field of other atoms or molecules. The first photoassociation experiments in a lattice were done with homonuclear Rb_2 molecules [19,20]. Polar KRb molecules have been produced in an optical lattice by using magnetic-dipole transitions [12].

In this Letter we propose an approach to create polar molecules by using a microwave field. Molecules can be produced by a single electric-dipole transition in excited vibrational states of the ground configuration as schemati-

cally shown in Fig. 1. For this proposal we will use the unique property of polar molecules, the existence of a nonzero permanent dipole moment, which allows electric-dipole transitions from trap levels of optically confined atoms to excited rovibrational levels or between rovibrational levels of the ground electronic potentials [15]. We believe the proposed method has advantages over other association methods. First of all, it explores transitions within the vibrational levels of the ground-state potential; therefore, it does not rely on the structure and lifetime of the excited states potential as it does for the photoassociation (PA) method. Second, it is not restricted to atomic systems that have magnetic Feshbach reso-

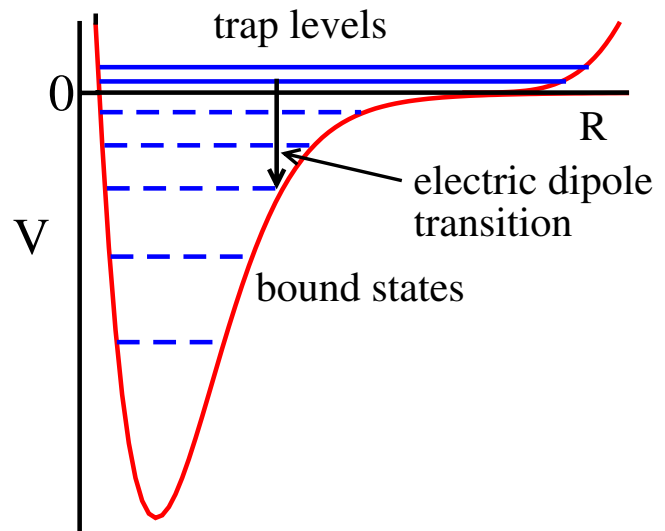


FIG. 1 (color online). Association scheme to create polar molecules with microwave fields. The schematic drawing shows the ground-state potential plus a trapping potential as a function of interatomic separation R . The nonzero permanent dipole moment allows electric-dipole transitions between trap levels and vibrational levels of the ground-state potential.

nances. On the other hand, our proposal can only be used with heteronuclear molecules, which possess the permanent dipole moments. There are, however, many such molecules. Moreover, we might envision binding molecules together by microwave-frequency electric-dipole transitions to create even bigger molecules or making production of molecules spatially dependent by applying additional spatially dependent static electric or magnetic fields.

The bottleneck in any molecule forming process from its constituent atoms is the transition from two free atoms to a weakly bound “floppy” molecule. As we will show later, the molecular production rates are such that our method as well as any other association method will benefit from confining the atoms in an optical lattice.

In this Letter we focus on the detailed numerical verification of the proposed association scheme for KRb and RbCs. We assume that two atoms of different species are confined in sites of an optical lattice and prepared in the lowest motional state of these sites. For simplicity, we assume that this confinement can be described by a spherically symmetric harmonic trap with frequency ω . This also leads to a harmonic trapping potential for the relative motion between atoms. Such a trapping potential must be added to and modifies the long-range behavior of the molecular potentials. For our calculation, the frequency of the trapping potential is set to $\omega/2\pi = 200$ kHz. The lowest harmonic oscillator levels are drawn schematically as the solid lines in Fig. 1.

We use a microwave field to induce a electric-dipole transition and to associate the lowest and rotationless trap level into a weakly bound vibrational level with one unit of rotational angular momentum of the $a^3\Sigma^+$ or $X^1\Sigma^+$ state. The transition moment is determined by the matrix element

$$d = C \int_0^\infty dR \phi_{v,\ell=1}(R) d(R) \phi_{\text{trap},\ell=0}(R), \quad (1)$$

where R is the interatomic separation and $d(R)$ is the permanent electronic dipole moment of the $a^3\Sigma^+$ or $X^1\Sigma^+$ states of KRb and RbCs obtained from Refs. [21,22]. The wave function $\phi_{\text{trap},\ell=0}$ is the lowest harmonic oscillator state of either the $a^3\Sigma^+$ or $X^1\Sigma^+$ potential plus a harmonic potential. The function $\phi_{v,\ell=1}$ is a $\ell = 1$ or p -wave rovibrational wave function of the same ground-state potential as the initial state with binding energy $E_{v\ell}$. The dimensionless coefficient C of the order of 1 contains the angular dependence of the transition and, in principle, depends on the magnetic sublevels as well as the polarization of the microwave photon. Here we assume it is equal to 1. Furthermore, for the purposes of this study, the hyperfine interaction and relativistic splitting between the $\Omega = 0^-$ and 1 components of the $a^3\Sigma^+$ potential can be neglected. For an accurate prediction of binding energies of the ground-state potentials we needed to combine the best electronic potentials available [22,23], RKR data [24–26], and long-range dispersion coefficients [27,28].

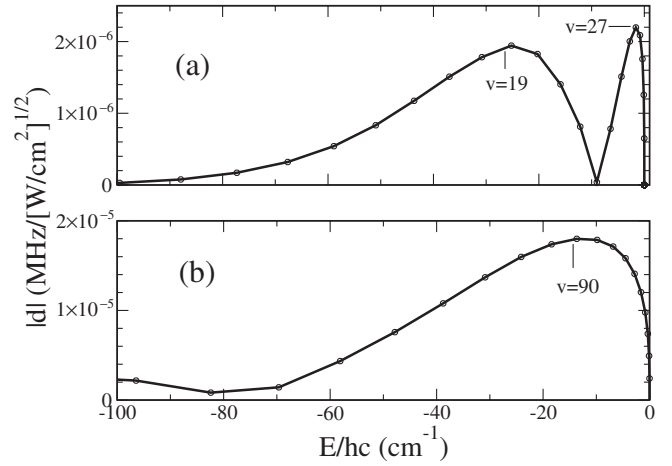


FIG. 2. The vibrationally averaged dipole moment of KRb for transitions from the first trap level of a harmonic potential with a frequency of $\omega/2\pi = 200$ kHz to vibrational levels of the $a^3\Sigma^+$ state [panel (a)] and $X^1\Sigma^+$ state [panel (b)] as a function of binding energy of the final state.

The theoretical electronic potentials have been slightly modified to fit to experimental measurements of scattering lengths where available [29,30].

Figures 2 and 3 show the absolute value of the transition dipole moment from the first trap level to the vibrational levels of the same potential for KRb and RbCs, respectively. The horizontal axis corresponds to the binding energies of the final state, which can also be interpreted as the microwave frequency needed to make the transition from the trap level. The markers on the curves in the figures correspond to bound vibrational states of the potentials. Uncertainties in the binding energies will not affect the functional shape of the transition dipole moment. In Fig. 2 the maximum association dipole moment for the $a^3\Sigma^+$ potential occurs for vibrational levels $v = 27$ and $v = 19$

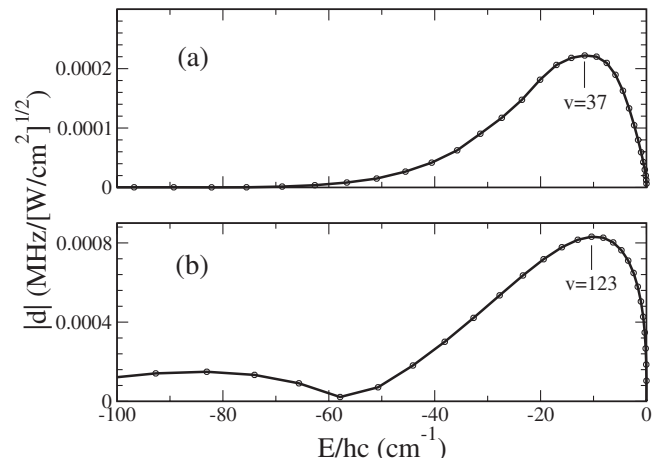


FIG. 3. The vibrationally averaged dipole moment of RbCs for transitions from the first trap level of a harmonic potential with a frequency of $\omega/2\pi = 200$ kHz to vibrational levels of the $a^3\Sigma^+$ state [panel (a)] and $X^1\Sigma^+$ state [panel (b)] as a function of binding energy of the final state.

bound by about -1.5 and -25 cm^{-1} , respectively, and for vibrational level $\nu = 90$ bound by about -13 cm^{-1} for the $X^1\Sigma^+$ potential. In Fig. 3 the maximum association dipole moment occurs for the $\nu = 37$ and $\nu = 123$ vibrational levels at a binding energy of approximately -10 cm^{-1} for both the $a^3\Sigma^+$ and $X^1\Sigma^+$ potentials, respectively.

The vibrationally averaged dipole moments are not the only relevant quantities for determining a successful association mechanism. Once a molecule has been formed the same microwave field can transfer it into trap levels other than the lowest. This could limit the number of molecules that can be made. However, in strongly confined traps this effect can be made small. For field intensities below $I = 10$ kW/cm^2 the dipole matrix element of the microwave transition, which is in units of MHz given by $V/h = d(\text{MHz}/\sqrt{W/\text{cm}^2}) \times \sqrt{I(W/\text{cm}^2)}$, is smaller than 100 kHz and thus less than our harmonic trapping frequency of $\omega/2\pi = 200$ kHz. As a result we will have a perfect two level system. This will prevent loss of molecules due to population of higher trap levels during Rabi oscillations.

We also find that the transition dipole is proportional to $\omega^{3/4}$, or, alternatively, as inversely proportional to the square root of the volume of the trap state. A tighter trap will increase the transition dipole moment. The proportionality is consistent with the Wigner threshold behavior for vibrationally averaged transition dipole moments. In other words, association of molecules in optical lattices is favorable.

References [22,31] have shown that the room-temperature black-body and natural lifetime of vibrational levels of the $a^3\Sigma^+$ and $X^1\Sigma^+$ states of KRB and RbCs are at least as large as 10^6 s. The large lifetime of these vibrational levels allows us sufficient time to create a dense cloud of molecules in an optical lattice.

The dipole moments in Figs. 2 and 3 are rather small compared to the dipole moments of first step of optical photoassociation. The latter correspond to transitions from a trap level to high vibrational levels of electronically excited potentials [32]. However, this is only half of this molecular formation process. Often the second step consists of a spontaneous decay into many rotational and vibrational levels of the ground-state potential. Our estimate of this process shows that the total transition dipole moment has the same order of magnitude as our microwave process.

It is often preferable for molecules to be vibrationally cold. We propose that the microwave transition will be followed by one optical Raman transition to form the vibrationally cold molecules. Hence, we apply a microwave field to accumulate a sufficient number of molecules in one excited vibrational level of either the a or X potential, and then use an optical Raman transition to convert these excited molecules into vibrationally cold $\nu = 0$, $\ell = 1$ $X^1\Sigma^+$ molecules. To show the strength of this scheme we calculate the effective dipole moment for the two-photon

optical Raman transition, focusing on the $\nu = 27$ $\ell = 1$ and $\nu = 37$ $\ell = 1$ vibrational levels of the $a^3\Sigma^+$ potential of KRB and RbCs, respectively, as starting levels of this process. These vibrational levels are chosen as they have the largest Rabi matrix elements with the lowest trap level. The $\nu = 37$ level of RbCs was used in experiment [17] to produce the $\nu = 0$ $X^1\Sigma^+$ molecules. The effective dipole moment for the two-photon Raman transition is $d_{\text{eff}} = C'd_1d_2/(\Delta + i\gamma/2)$, where d_1 and d_2 are the vibrationally averaged transition dipole moments for the upwards and downwards transition, respectively. In this equation the dipole moments have units of $\text{MHz}/\sqrt{W/\text{cm}^2}$. The quantities Δ and γ in MHz are the detuning from and the linewidth of a rovibrational level of the intermediate state. Finally, the dimensionless coefficient C' contains all angular momentum information and is assumed to be 1.

The results of our calculation are shown in Figs. 4(a) and 4(b). The vertical axis gives the product of the absolute value of the upward and downward transition dipole and the horizontal axis shows the binding energies of intermediate states relative to their own dissociation limit. In both cases we have used the vibrational levels of the 3(1) and 4(1) potentials as intermediate states as these are good candidates for molecular formation [10,17]. A comparison of the transition rates determined from Fig. 4 with the more conventional photoassociation scheme of the two-photon Raman transition from the trap level shows more than a 3 order of magnitude increase.

We also propose to apply a single microwave electric-dipole transition to Feshbach molecules in order to convert these molecules into more deeply bound states. Feshbach

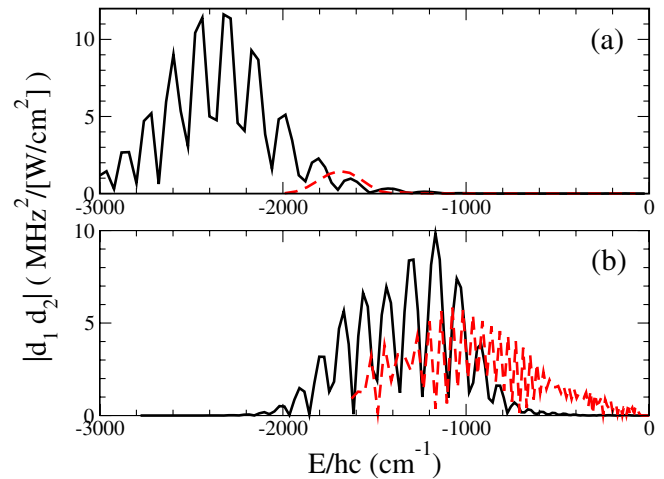


FIG. 4 (color online). The two-photon vibrationally averaged dipole moments of KRB [panel (a)] and RbCs [panel (b)] for the optical Raman transition as a function of vibrational binding energies of the 3(1) (solid line) and 4(1) (dashed line) intermediate states. The panel (a) corresponds to the pathway from $\nu = 27$ of the $a^3\Sigma^+$ potential to the $\nu = 0$, $\ell = 1$ $X^1\Sigma^+$ ground state of KRB. The panel (b) corresponds to the pathway from $\nu = 37$ of the $a^3\Sigma^+$ potential to the $\nu = 0$, $\ell = 1$ $X^1\Sigma^+$ state of RbCs.

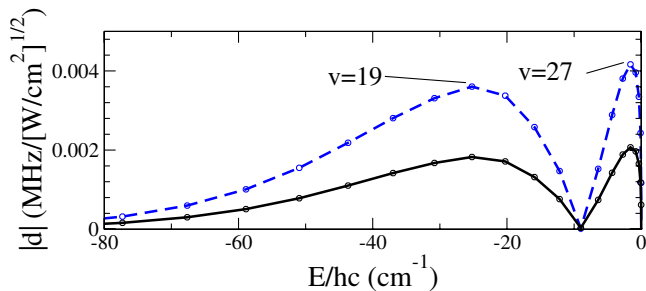


FIG. 5 (color online). The vibrationaly averaged dipole moment for the transition from the $\ell = 0$, $v = 31$ (solid line) and $\ell = 0$, $v = 30$ (dashed line) levels of the $a^3\Sigma^+$ of KRb to more deeply bound $\ell = 1$ vibrational levels of this state as a function of binding energy of the final state.

molecules were successfully created in KRb [29,33], while so far none have been observed in RbCs. The molecular level is bound by no more than 0.1 cm^{-1} , a typical atomic hyperfine energy. In fact, this bound state is often the last or second from the last bound state of the a or X state. Figure 5 shows results of a calculation of the transition dipole moment from the last and second from the last vibrational levels of the $a^3\Sigma^+$ potential of KRb to other vibrational levels. A comparison of Figs. 2 and 5 shows a significant increase in the dipole moment.

The range of microwave frequencies, considered in Figs. 2, 3, and 5 is only easily available for wave numbers less than 2 cm^{-1} . For these frequencies the relevant dipole moments are approximately half of their maximal values. Of course, use of more polar molecules increases these dipole moments significantly. The technology, which creates radiation between 10 and 30 cm^{-1} , where the transition dipole moment is maximal, is now being developed for spectroscopy of large biologically relevant molecules [34].

In this Letter we have searched for routes to simple and controllable production of molecules in tightly confining optical traps and optical lattices. As we have shown, a microwave-driven molecular association method, which is unique to polar molecules, is one such option. This method does not suffer from uncontrollable spontaneous decay from the excited states. We also have shown that molecules created by microwave transitions are easily converted to the vibrationally cold molecules.

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