Interaction-induced ferroelectricity in the rotational states of polar molecules

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We show that a ferroelectric quantum phase transition can be driven by the dipolar interaction of polar molecules in the presence a microwave field. The obtained ferroelectricity crucially depends on the harmonic confinement potential, and the macroscopic dipole moment persists even when the external field is turned off adiabatically. The transition is shown to be second order for fermions and for bosons of a smaller permanent dipole moment, but is first order for bosons of a larger moment. Our results suggest the possibility of manipulating the microscopic rotational state of polar molecules by tuning the trap's aspect ratio (and other mesoscopic parameters), even though the later's energy scale is smaller than the former's by six orders of magnitude.

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The successful realization of high phase-space-density polar molecules in Ni et al. [1] opens a new direction of strongly correlated quantum gases: ultracold polar molecules. Many experimental [2] and theoretical works [3–6] have been carried out in recent years. Most many-body physics proposed so far are based on the situation when the intermolecule effective interaction is predetermined (by a separated calculation) [7] and cannot be affected by the many-body physics. On the other hand, the reverse operation, that is, manipulating the molecular rotational state by changing a mesoscopic parameter (say trapping frequency or particle number, etc.), is still unfeasible, because the rotational energy is about six orders of magnitude larger than the interaction energy in a dilute gas. This makes systems of polar molecules very different from spinor atoms [8], although they both have rich internal states and long-ranged dipolar interaction. For the same reason, a spontaneous long-ranged order of molecular rotational state seems impossible either [9], while the related many-body phenomena, like (anti-)ferromagnetism and multiferroics [10], have been important subjects of solid state physics for decades.

In this article, we demonstrate that the rotational state of polar molecules can be manipulated by mesoscopic parameters through an external AC field [3]: When the trapping potential is elongated and the field frequency is close to the rotational energy [see Fig. 1(a)], the molecular rotational state is "locked"



FIG. 1. (Color online) (a) The lowest two rotational eigenstates of a polar molecule. Here we consider a linearly polarized AC field to couple state ($|g\rangle = |0, 0\rangle$) and state ($|e\rangle = |1, 0\rangle$) with detuning Δ . (b) Schematic figure for the ferroelectricity in the presence of a microwave field. The blue (up-down) arrows indicate the oscillating dipole moments.

by interaction, leading to a spontaneous ferroelectric order. Different from the standard dressed state of a single molecule, the interaction-induced macroscopic dipole moment here sustains even when the external field is turned off [6]. We calculate the quantum phase diagrams for both bosonic and fermionic polar molecules in an elongated ellipsoid trap. We then discuss the nature of the phase transition and how it can be tuned via mesoscopic parameters, like trap's aspect ratio or the detuning of an external field.

The system Hamiltonian of N polar molecules includes both the rotational and the orbital degrees of freedom: $H_{\text{tot}} =$ $\sum_{i}^{N} H_{\text{rot},i} + H_{\text{orb}}$. Here, $H_{\text{rot},i} = B \mathbf{J}_{i}^{2} - \mathbf{d}_{i} \cdot \mathbf{E}(t)$ describes the rotational state of the *i*th molecule with J_i and *B* being the angular momentum operator and the rotational constant, respectively. $\mathbf{E}(t)$ is the external field, and \mathbf{d}_i is the electric dipole moment. The orbital Hamiltonian is $H_{\text{orb}} \equiv \sum_{i=1}^{N} \left[\frac{\mathbf{p}_{i}^{2}}{2m} + V_{\text{t}}(\mathbf{r}_{i})\right] + \frac{1}{2} \sum_{i\neq j}^{N} \left[\frac{\mathbf{d}_{i} \cdot \mathbf{d}_{j} - 3(\mathbf{d}_{i} \cdot \mathbf{e}_{ij})(\mathbf{d}_{j} \cdot \mathbf{e}_{ij})}{|\mathbf{r}_{i} - \mathbf{r}_{j}|^{3}}\right]$, where \mathbf{r}_{i} and \mathbf{p}_{i} are the variables and \mathbf{r}_{i} and \mathbf{r}_{i} are the position and momentum operators, and m is the molecule mass. N is the total number of particles and $V_t(\mathbf{r}) =$ $\frac{1}{2}m[\omega_{\rho}^{2}(x^{2}+y^{2})+\omega_{z}^{2}z^{2}]$ is the external trapping potential. The second term is the dipolar interaction with $\mathbf{e}_{ii} \equiv \mathbf{r}_{ii}/|\mathbf{r}_{ii}|$ being the unit vector of $\mathbf{r}_{ij} \equiv \mathbf{r}_i - \mathbf{r}_j$. Following the spirit of the mean-field approximation, without losing generality, we can approximate the ground-state wave function by a factorizable form: $\Psi(1, 2, ..., N) = \Phi_{\text{orb}}(\theta; \mathbf{r}_1, ..., \mathbf{r}_N) \otimes \prod_{i=1}^{N} |\theta_i\rangle_i$, where $\Phi_{\text{orb}}(\theta; \mathbf{r}_1, ..., \mathbf{r}_N)$ is the orbital wave function with a variational parameter, θ_i , and $|\theta_i\rangle_i$ describes the rotational state of the *i*th particle, as defined below.

In this article, we consider the situation when a linearly polarized AC, $\mathbf{E}(t) = E_{AC} \cos(\omega t)\hat{z}$, is applied to couple rotational states $|g\rangle = |0, 0\rangle$ and $|e\rangle = |1, 0\rangle$ [see Fig. 1(a)]. Solving the Floquet equation [12] within the rotating wave approximation, we obtain the following time-independent Hamiltonian:

$$\tilde{H}_{\text{rot},i} = -\frac{\hbar\Delta}{2} - \frac{\hbar\Delta_{\Omega}}{2} \begin{bmatrix} \cos\alpha & \sin\alpha\\ \sin\alpha & -\cos\alpha \end{bmatrix}, \quad (1)$$

in the basis of { $|g\rangle$, $e^{-i\omega t}|e\rangle$ }, where $\Omega \equiv d_0 E_{AC}/\hbar$ is the Rabi frequency, $\Delta \equiv \omega - 2B/\hbar$ is the detuning, $\Delta_{\Omega} \equiv \sqrt{\Delta^2 + \Omega^2}$, and $\tan \alpha \equiv \Omega/\Delta$. Here $d_0 \equiv |\langle g|\mathbf{d}_i|e\rangle|$ is the permanent dipole moment. A general total state can be expressed to be $|\theta_i\rangle_i = \cos(\frac{\theta_i}{2})|v_+\rangle e^{-iE_+t/\hbar} + \sin(\frac{\theta_i}{2})|v_-\rangle e^{-iE_-t/\hbar}$, where $E_{\pm} = \frac{-\hbar}{2} (\Delta \pm \sqrt{\Delta^2 + \Omega^2})$ are the eigenenergies, and $|v_+\rangle =$ $-\sin(\alpha/2)|g\rangle + \cos(\alpha/2)e^{-i\omega t}|e\rangle$ and $|v_{-}\rangle = \cos(\alpha/2)|g\rangle +$ $\sin(\alpha/2)e^{-i\omega t}|e\rangle$ are the corresponding eingenstates of $H_{\text{rot},i}$. After a straightforward calculation, we can obtain the rotational energy per particle, $E_{\rm rot}(\theta)/N = -\frac{h\Delta}{2}$ – $\frac{\hbar\Delta_{\Omega}}{2}\sum_{i=1}^{N}\cos\theta_{i}$, and the averaged dipole moment (in the limit of small Δ_{Ω}/ω : $\langle \mathbf{d}_i(t) \rangle \equiv \langle_i \theta_i | \mathbf{d}_i | \theta_i \rangle_i = d_0 \sin(\theta_i - \theta_i)$ α) cos(ωt) \hat{z} , which is oscillating in time. If $\hbar \Delta_{\Omega}$ is larger than orbital energy scales, the rotational energy is then always minimized at a single dressed state, $|v_{+}\rangle$ (i.e., $\theta_{i} = 0$) with a finite oscillating dipole moment. However, this state alone is not a true ferroelectric state [9], because the dipole moment disappears in the limit of zero external field (i.e., $\alpha \rightarrow 0$). This situation, however, can be totally different when Δ is tuned to be comparable to the interactional energy, as shown in more details below.

Before further investigation, we note that the θ_i , in principle, can depend on the confinement geometry: when the molecule cloud is elongated along the *z* axis (i.e., the polarization direction), θ_i will tend to be uniform in space in order to gain the interaction energy. However, if the cloud is oblate in the *x*-*y* plane, θ_i can become nonuniform in space, that is, forming a domain wall structure or a rotation-orbital texture [16]. Since here we concentrate on the ferroelectric order rather than the complete phase diagram, we will always consider a highly elongated trap and assume a uniform polarization for simplicity.

With such approximation, the interaction strength can be characterized by a dipolar length scale:

$$a_d(\theta) \equiv \frac{m \langle \langle \mathbf{d}(t) \rangle \cdot \langle \mathbf{d}(t) \rangle \rangle_t}{\hbar^2} = \frac{a_d^{(0)}}{2} \sin^2(\theta - \alpha), \quad (2)$$

where $a_d^{(0)} \equiv m d_0^2 / \hbar^2$, and $\langle, \ldots, \rangle_t$ is the time average. To study its effect on the orbital many-body wave function, we first consider bosonic molecules at zero temperature, where the orbital condensate wave function can be well approximated by a Gaussian type form: $\Psi(\mathbf{r}) = \frac{N^{1/2}}{\pi^{1/4} R_\rho R_z^{1/2}} e^{-(\rho^2 + z^2/\beta^2)/R_\rho^2}$ with the horizontal radius R_ρ and the aspect ratio β . The orbital condensate energy, $E_{\text{orb}}^{\text{BEC}}(\theta)$, is [13,14]

$$\frac{E_{\text{orb}}^{\text{BEC}}(\theta)}{N\hbar\omega_{\rho}/2} = \frac{a_{\rho}^{2}}{R_{\rho}^{2}} \left(1 + \frac{1}{2\beta^{2}}\right) + \frac{R_{\rho}^{2}}{a_{\rho}^{2}} \left(1 + \frac{\beta^{2}}{2\kappa^{2}}\right) + \frac{2Na_{s}a_{\rho}^{2}}{\sqrt{2\pi}\beta R_{\rho}^{3}} + \frac{16A_{2}(\beta)}{3\sqrt{10\pi}} \frac{Na_{d}(\theta)a_{\rho}^{2}}{R_{\rho}^{3}}, \quad (3)$$

where $a_{\rho} = \sqrt{\hbar/m\omega_{\rho}}$ is the oscillator length, and a_s is the *s*-wave scattering length. $\kappa \equiv \omega_{\rho}/\omega_z$.

Now we minimize the total energy, $E_{\text{tot}}^{\text{BEC}}(\theta) = E_{\text{rot}}(\theta) + E_{\text{orb}}^{\text{BEC}}(\theta)$, with respect to variational parameters, R_{ρ} , β , and θ . For an elongated trap (i.e., $\beta \gg 1$), we can use $A_2(\beta) \rightarrow \frac{-\sqrt{5}}{8\beta}$ and therefore the interaction energy term of Eq. (3) becomes the same as the result of a nondipolar condensate with an effective length, $a_{\text{eff}}(\theta) \equiv a_s - a_d(\theta)/3$. Within the Thomas-Fermi limit (i.e., $N \gg 1$ and hence the kinetic energy is negligible), the minimum energy at a given θ can be calculated analytically [15]. We obtain $\beta = \kappa \equiv \omega_{\rho}/\omega_z$, and

$$\frac{E_{\rm orb}^{\rm BEC}(\theta)}{N} = \frac{5}{4} \left(\frac{2}{\pi}\right)^{1/5} \hbar \bar{\omega} \left(\frac{N a_{\rm eff}(\theta)}{\bar{a}}\right)^{2/5},\qquad(4)$$

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where $\bar{\omega} \equiv (\omega_{\rho}^2 \omega_z)^{1/3}$ and $\bar{a} \equiv \sqrt{\hbar/m\bar{\omega}}$. It is easy to see that the system becomes unstable toward collapse when $a_{\text{eff}}(\theta) < 0$, giving the same condition derived in magnetic atoms [14]. Taking zero field limit and expanding $E_{\text{tot}}^{\text{BEC}}(\theta)$ to the two leading orders of small θ , we have

$$\frac{E_{\text{tot}}^{\text{BEC}}(\theta)}{N} \approx \left(\frac{\hbar|\Delta|}{4} - \chi_b\right)\theta^2 + \left(\lambda\chi_b - \frac{\hbar|\Delta|}{48}\right)\theta^4, \quad (5)$$

after shifting the total energy to zero at $\theta = 0$. Here we define $\chi_b \equiv \frac{a_d^{(0)}}{15a_s} \chi_b^0$ to be the "electric susceptibility" and $\lambda \equiv \frac{1}{3} - \frac{a_d^{(0)}}{20a_s}$. Here $\chi_b^0 \equiv E_{orb}^{BEC}(0)N^{-1}$. It is easy to see that a second-order quantum phase transition from a paraelectricity toward a ferroelectricity can take place when $|\Delta| < \Delta_c \equiv 4\chi_b/\hbar$ and when $\lambda\chi_b - \frac{\hbar\Delta_c}{48} > 0$ (i.e., $a_d^{(0)} < 5a_s$). In Fig. 2(a), we show $E_{tot}^{BEC}(\theta)$ as a function of the variational parameter, θ , near the phase transition of $a_d^{(0)}/a_s = 2$. However, when $a_d^{(0)} > 5a_s$, the coefficient of the θ^4 term becomes negative, leading to a first-order phase transition instead [see Fig. 2(b)]. In Fig. 2(c), we show the calculated quantum phase diagram for such elongated condensate, as a function of $a_d^{(0)} \sim 1.33$. When $a_d^{(0)}/a_s > 6$, the condensate becomes collapsed due to the strong attractive dipolar interaction. We can further derive the



FIG. 2. (Color online) Results of bosonic polar molecules. (a) Total ground-state energy as a function of θ for $a^{(0)}/a_s = 2$. Curves from top to bottom are, respectively, for $\hbar \Delta / \chi_b^0 = 0.6$, 0.5, 0.4, and 0.3. (b) The obtained effective dipolar length, $a_d(\theta)$, as a function of the detuning, Δ . Curves are for $a_d^{(0)}/a_s = 2$, 4, and 5.5, respectively, from left to right. (c) Quantum phase diagram as a function of $a_d^{(0)}$ and Δ . The para- to ferroelectricity transition is second order for $a_d^{(0)}/a_s < 5$, while it is first order for $a_d^{(0)}/a_s > 5$. When $a_d^{(0)}/a_s > 6$, the condensate becomes unstable toward collapse. (d) Critical detuning for the second-order transition line as a function of trapping aspect ratio, κ . Curves from top to bottom are for $a_d^{(0)}/a_s = 5$, 4, and 3. Note that we have taken $\Omega \to 0$ for all results shown here.

critical detuning as a function of the trap's aspect ratio, $\kappa = \omega_{\rho}/\omega_z$: $\hbar\Delta_c = \frac{4a_d^{(0)}}{15a_s}\chi_b^0 = \frac{4a_d^{(0)}}{15a_s}\chi_b^0(\kappa = 1)\kappa^{-2/5}$, and therefore the ferroelectricity can be also tuned easily by changing the trap's aspect ratio as shown in Fig. 2(d). Note that the decay of Δ_c for larger κ is simply due to the reduced particle density. Taking ³⁹K⁸⁷Rb as an example, we have $d_0 = 0.5$ Debye and hence $a_d^{(0)} = 0.47 \ \mu$ m. Using $a_s = 0.2 \ \mu$ m, we can easily obtain $\Delta_c = 2\pi \times 250$ Hz for $N = 10^4$, $\omega_{\rho} = 2\pi \times 100$ Hz, and $\kappa = 100$.

Now we turn to systems of fermionic molecules. However, different from the bosonic case, the assumption of uniform polarization may fail if Fermi energy is larger than the energy gap Δ_{Ω} [see Eq. (1) and the text], unless the system is in a strongly interacting regime with dipole moment oscillating along the elongated axis [see Fig. 1(b)]. This condition is consistent with the ferroelectric order we investigate in this article, but may lead to system collapse. Here, for simplicity, we will first assume a uniform ferroelectricity for the fermionic case, and will then numerically evaluate relevant conditions in more detail later. However, even for a uniform static field, only a variational study [19]and Fermi-liquid theory in a uniform system [18] are studied in the literature. It is therefore worthy to show our improved calculation on this aspect first, using a full self-consistent Hartree-Fock theory with the local density approximation (SCHF+LDA). The obtained orbital energy is

$$E_{\text{orb}}^{\text{FL}}(\theta) = \int_{\mathbf{r},\mathbf{k}} \left[\epsilon_{\mathbf{k}}^{0} + V_{\text{t}}(\mathbf{r}) + \frac{1}{2} \Sigma(\mathbf{k},\mathbf{r}) \right] f_{\mathbf{k}}(\mathbf{r}), \quad (6)$$

where $\Sigma(\mathbf{k}, \mathbf{r}) = \int_{\mathbf{r}', \mathbf{k}'} [V_d(\mathbf{r} - \mathbf{r}') - \tilde{V}_d(\mathbf{k} - \mathbf{k}')\delta(\mathbf{r} - \mathbf{r}')] \times$ $f_{\mathbf{k}'}(\mathbf{r}')$ is the static local self-energy, and $f_{\mathbf{k}}(\mathbf{r}) \equiv \{\exp[(\epsilon_{\mathbf{k}}^{0} + V_{t}(\mathbf{r}) + \Sigma(\mathbf{k}, \mathbf{r}) - \mu)/k_{B}T] + 1\}^{-1}$ is Fermi distribution at temperature $T \cdot \int_{\mathbf{r},\mathbf{k}} = \iint \frac{d\mathbf{r}d\mathbf{k}}{(2\pi)^{3}}$ is the integration over the whole phase space. $V_d(\mathbf{r}) = |\langle \mathbf{d} \rangle|^2 (r^2 - 3z^2) |\mathbf{r}|^{-5}$ is dipolar interaction and $\tilde{V}_d(\mathbf{k})$ is its Fourier transform. $\epsilon_{\mathbf{k}}^{0} = \hbar^{2} \mathbf{k}^{2} / 2m$, and μ is the chemical potential. Equation (6) can also be applied to systems of finite temperature, but in this article we just consider the zero-temperature situation. In Fig. 3(a), we show the calculated density distribution for different aspect ratios of the trap, and in (b) and (c) we show the obtained cloud's aspect ratio (normalized by the noninteracting aspect ratio). Here $R_z \equiv (\frac{1}{N} \int_{\mathbf{r},\mathbf{k}} f_{\mathbf{k}}(\mathbf{r}) z^2)^{1/2}$ and $K_z \equiv (\frac{1}{N} \int_{\mathbf{r}, \mathbf{k}} f_{\mathbf{k}}(\mathbf{r}) k_z^2)^{1/2}$ are the averaged radii in the spatial and momentum space (similar definitions for R_x and K_x). Our results are consistent with Ref. [19] in the spatial aspect ratio, but disagree with them in the momentum aspect ratio. It is probably because their variational approach did not capture the full contribution of Hartree energy.

From Eq. (6), one can see that the susceptibility has two contributions: one is from the self-energy in Eq. (6), and the other is from the single-particle energy via the deformation of the Fermi surface. In the low- temperature limit, the latter contribution can be neglected, because it involves only a surface integral of the Fermi sea in the phase space. Besides, most effects on the self-energy in $f_{\mathbf{k}}(\mathbf{r})$ is reduced by shifting the chemical potential due to the particle number conservation. As a result, we can just keep the interaction effect to the leading order of $a_d(\theta)$, which is proportional to $|\langle \mathbf{d} \rangle|^2$ in $V_d(\mathbf{r})$. The first-order transition does not exist here for the similar

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FIG. 3. (Color online) (a) Zero temperature density profile (after column integration) for fermionic polar molecules, ${}^{40}K^{87}Rb$, in three different trap's aspect ratios, κs . Here we use $N = 3 \times 10^5$, $|\langle \mathbf{d} \rangle| = 0.5$ Debye, and a fixed $\omega_{\rho} = 2\pi \times 100$ Hz. (b) and (c) are renormalized cloud's aspect ratios in the spatial and momentum density distribution as a function κ . $|\langle \mathbf{d} \rangle| = 0.1$, 0.3 and 0.4 Debye from bottom to top.

reason. After rescaling the length scale to $r_F \equiv \sqrt{2\epsilon_F^{\text{ho}}/m\omega_x^2}$ and the momentum scale to $k_F \equiv \sqrt{2m\epsilon_F^{\text{ho}}/\hbar^2}$ with $\epsilon_F^{\text{ho}} \equiv (6N)^{1/3}\hbar\bar{\omega}$ [15], we obtain (taking $\Omega \to 0$), $E_{\text{orb}}^{\text{FL}}(\theta)N^{-1} = E_{\text{orb}}^{\text{FF}}N^{-1} - 48\sqrt{3N}\sin^2\theta(d_0^2/\bar{a}^3)C(\kappa)$, where $E_{\text{orb}}^{\text{FF}}$ is the energy of free fermions. $C(\kappa) \equiv \frac{4}{\kappa}\sqrt{\frac{\pi}{5}}\int_{\mathbf{r},\mathbf{k}}'\int_{\mathbf{r},\mathbf{k}}' |Y_{20}(\hat{\mathbf{e}}_r)|\mathbf{r} - \mathbf{r}'|^{-3} + \frac{4\pi}{3}Y_{20}(\hat{\mathbf{e}}_k)\delta(\mathbf{\tilde{r}} - \mathbf{\tilde{r}}')]$ is a dimensionless function of κ with $Y_{lm}(\hat{\mathbf{e}}_{r,k})$ being the spherical harmonics. $\hat{\mathbf{e}}_k$ is the unit along $\mathbf{k} - \mathbf{k}'$. $\int_{\mathbf{r},\mathbf{k}}' z^2/\kappa^2 + \mathbf{\tilde{k}}^2 = 1$. Note that $C(\kappa)$ plays a similar role as $A_2(\beta)$ in Eq. (3). Therefore, expanding the total energy to the quadratic order of θ , we can obtain the "susceptibility" per particle to be $\chi_f \equiv \chi_f^0 C(\kappa)$, where $\chi_f^0 \equiv 48(3N)^{1/2}d_0^2/\bar{a}^3$. The critical detuning for ferroelectricity is given by $\Delta_c = 4\chi_f/\hbar$ as shown



FIG. 4. (Color online) Results of fermionic polar molecules. (a) Quantum phase diagram in terms of the trap's aspect ratio, κ , and the detuning Δ , in the zero field limit. (b) The calculated dipolar length, $a_d(\theta)$, as a function of Δ for $\kappa = 20$. Results for different Rabi frequencies are shown together.

in Eq. (5). In Figs. 4(a) and 4(b) we show the calculated quantum phase diagram, and the dipolar strength as a function of detuning.

As mentioned earlier, to justify our theory, we have to satisfy three conditions all together: (i) unform polarization, (ii) stability against collapse, and (iii) ferroelectric order. Qualitatively speaking, to satisfy condition (i), one needs the sum of energy gap $(\hbar \Delta)$ and interaction energy to be larger than the Fermi energy, while for (ii) one needs larger kinetic energy than the interaction energy. Since $\hbar \Delta_c$ is basically the same as the interaction energy, we find these three conditions can be satisfied (qualitatively) if $\hbar \Delta_c$ is just slightly smaller than $\varepsilon_F^{\text{ho}}$. Putting experimental parameters, we find that ⁴⁰K⁸⁷Rb is not a good candidate for ferroelectricity, since its permanent dipole moment is still too small. However, for ${}^{6}\text{Li}{}^{19}\text{F}$ molecules ($d_{0} =$ 6.5 Debye) with $N = 10^4$, $\omega_\rho = 2\pi \times 100$ Hz, and $\kappa = 100$, we have $C(\kappa) = 3 \times 10^{-4}$ and hence $\Delta_c = 4\chi_f/\hbar \sim 786$ Hz, very close to the Fermi energy $\varepsilon_F^{\rm ho} \sim 843$ Hz. Therefore, it is reasonable to believe that the system is still stable against

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collapse and has a uniform polarization in the ferroelectric phase. A full calculation including species mixtures will be necessary for a more quantitative study.

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In conclusion, we show that an interaction-driven quantum phase transition from a paraelectric to a ferroelectric quantum gas can be realized in quantum gases of polar molecules. The ferroelectric order predicted here can be easily observed from the electric field distribution when the AC field is turned off adiabatically ($\alpha \rightarrow 0$). The resulting macroscopic polarization of rotational states can also be manipulated by changing the confinement's aspect ratio, *s*-wave scattering length (bosonic case), or detuning frequency. Such ferroelectric order cannot be generated in a single molecule picture.

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