Proposed Test of the Equivalence Principle with Rotating Cold Polar Molecules *

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We propose a novel scheme for testing the equivalence principle with rotating cold polar molecules whose angular momenta are polarized at different states. Molecules in specific rotational states are selected out via the hexapole state-selection technique and the gravitational acceleration g of molecules is measured by measuring the Doppler shift of the molecules free falling in the gravitational field. Some other possible methods of rotating molecules and measuring g are also mentioned. Molecules, as the test masses, have higher rotating speed and smaller dimension in comparison with mechanical gyroscopes and may open a new way for testing the equivalence principle and the possible coupling between rotation and gravity.

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It is well known that the equivalence principle, as one of the fundamental hypotheses of Einstein's general relativity, has been tested by many experiments,[1–5] but most of these experiments are with nonrotating test masses, treated as point masses. On the other hand, the spin-gravitational couplings of elementary particles have also been investigated for a long time in both theory and experiment.[6–10] In 2001, Zhang *et al.*[11] developed a phenomenological model for the spin-spin interaction between rotating extended bodies. In the model, a dimensionless parameter representing the strength of violation of the equivalence principle was defined as follows:

$$\eta_{\rm s} = \frac{\Delta g}{g} = \kappa\Big(\frac{\boldsymbol{S}_1 \cdot \boldsymbol{S}_{\rm e}}{Gm_1M_{\rm e}R_1} - \frac{\boldsymbol{S}_2 \cdot \boldsymbol{S}_{\rm e}}{Gm_2M_{\rm e}R_2}\Big), \qquad (1)$$

where G is the Newtonian gravitational constant, m_1, m_2, and $M_{\rm e}$ are the masses of the two rotating bodies and the Earth, respectively, and $\boldsymbol{S}_1$, $\boldsymbol{S}_2$, and $\boldsymbol{S}_{\rm e}$ are their spin angular momenta correspondingly, R_1 and R_2 are the distances between the centers of the two gyroscopes and the Earth, respectively, and the parameter κ represents the universal coupling factor for the spin-spin interaction. We have performed a series of double free fall experiments, in which the test masses are two gyroscopes with differing rotating senses, and no apparent differential acceleration has been observed at the relative level of 1.6×10^{-7}.[12]

The rotating speed of mechanical gyroscopes is usually limited by mechanical friction at the level of 10^4 rpm. To increase the rotating speed of the test masses, we propose a novel scheme for testing the equivalence principle by comparing the gravitational accelerations of rotating cold molecules whose angular momenta will be polarized vertically up and down, as shown in Fig. 1. Molecules with a particular rotational quantum number M which characterizes the projection of the total angular momentum onto a space-fixed axis means that their angular momenta are vertically polarized. For most molecules whose rotational constants at the order of GHz, molecules even in the lowest rotationally excited states have classical rotating speed of the order of 10^{12} rpm, which is 8 orders higher than that of mechanical gyroscopes. Therefore, molecules are more sensitive to test effects relative to the rotating speed. Testing the free fall of cold polar molecules with different polarized angular momentum states may open a new window for testing the equivalence principle.

To test the equivalence principle with rotating polar molecules, we should first prepare molecules with a particular angular momentum state, and then compare the free falling acceleration of cold molecules with different rotation states. To speed up the rotation frequency of molecules, a straightforward idea is to take advantage of the molecular dipole moment and use a microwave field[13] or a laser[14] to rotate molecules. The orientation of the molecular angular momentum will be determined by the rotational direction of the light field.

There is a relatively simple method to produce molecules in desired rotational states. As we know, molecules in thermal equilibrium have a rotational state distribution. If we can select molecules in a specific rotational state, then we can produce molecules with a particular angular momentum orientation. The hexapole state-selection technique can prepare molecules in a combination of rotational states, which can be further manipulated to get a single rotational state sensitive to the detecting light, and the theory and practice of state selection and focusing of symmetric-top molecules by electrostatic hexapole fields are well known.[15]

The first-order Stark effect of symmetric-top molecules is given by the following formula[16]

$$W^{(1)}_{\rm Stark} = -\frac{KM}{J(J+1)}\mu E, \qquad (2)$$

where μ is the molecular electric dipole moment, E is the electric field strength, J, K, and M are the

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usual rotational and projection quantum numbers of the symmetric-top molecule. For molecules in low-field-seeking states, i.e., $KM < 0$, they execute radial harmonic motion in the hexapole field, and molecules in a given state $|JKM\rangle$ will be focused at the same region. The rod voltage required for focusing a specific state $|JKM\rangle$ can be expressed as[15]

$$V_0 = -\frac{n^2\pi^2}{6}\frac{r_0^3}{l^2}\frac{mv^2}{\mu}\frac{J(J+1)}{KM}, \tag{3}$$

where n is the number of half-cycles of the molecular trajectory, r_0 is the hexapole radius, l is the length of the hexapole, m is the molecular mass, and v is the velocity of the molecular beam.

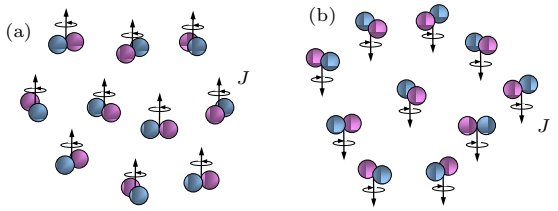

Fig. 1. The schematic diagram of rotating cold molecules whose angular momenta are polarized vertically up (a) and down (b) in the gravitational field of the Earth.

From Eq. (3), it is clear that the quantum numbers K and M play the same role. Thus the hexapole state-selection technique, which takes advantage of the molecular Stark effect, can only select a combination of rotational states which have a particular product of K and M for a given J. To further distinguish these different components of K and M, we propose to use a microwave field to manipulate molecular rotational states, and the specific method will be discussed in the following.

Table 1. The relative populations of several lowest rotational states $|JKM\rangle$ of CH_3F molecules which have a first-order Stark effect at the temperature of 5 K.

State	Population (%)
$\|11-1\rangle$ or $\|1-11\rangle$	1.62
$\|21-1\rangle$ or $\|2-11\rangle$	0.61
$\|21-2\rangle$ or $\|2-12\rangle$	0.61
$\|31-1\rangle$ or $\|3-11\rangle$	0.14
$\|31-2\rangle$ or $\|3-12\rangle$	0.14
$\|31-3\rangle$ or $\|3-13\rangle$	0.14

We take the fluoromethane (CH_3F) molecule as a sample of the test mass in our proposed experiment. As a symmetric-top molecule, CH_3F has a modest dipole moment (∼1.86 Debye), a relatively slight molecular weight (∼34.03) and rotational constants $A = 155.353$ GHz, $B = C = 25.5361$ GHz.[17] The relatively large dipole-to-mass ratio is helpful for the electrostatic Stark deceleration which will be mentioned below. Consider a supersonic CH_3F beam with a temperature of ∼5 K; molecules are mainly populated in the vibrational and electronic ground state and a few lowest rotational states. We calculate the relative populations of several lowest weak-field-seeking rotational states $|JKM\rangle$ which have a first-order Stark effect, and the results are listed in Table 1. Among these rotational states, the $|J = 1, K = \pm 1, M = \mp 1\rangle$ states have the largest population, and the relative population of each state is about 1.62%. For the sake of more molecules in the experiment, we would select out these rotational states via the electrostatic hexapole technique.

With a proper voltage on hexapole rods, molecules in two compositive rotational states, i.e., $|11-1\rangle$ and $|1-11\rangle$, can be selected, and then we should distinguish those two states. As we know, symmetric-top molecules in the rotational state $|JKM\rangle$ have rotational energy structure as follows:[16]

$$E_{J,K} = h[BJ(J+1) + (A-B)K^2]. \tag{4}$$

Therefore these two rotational states have the same rotational energy. However, we can apply a microwave field to remove this degeneracy. According to the selection rule, we could apply a σ^+ circularly polarized field to pump the $|1-11\rangle$ state to the $|2-12\rangle$ state or a σ^- circularly polarized field to pump the $|11-1\rangle$ state to the $|21-2\rangle$ state. We first discuss the case of σ^+ polarization, and the case of σ^- polarization is very similar. A bias static electrical field of 100 V/cm in the z direction is applied to split the different M levels due to the Stark effect. Then a σ^+ circularly polarized field also along the z direction is adjusted to resonance with the $|1-11\rangle$ to $|2-12\rangle$ transition, as shown in Fig. 2(a). The transition frequency is $4B$=102.145 GHz and thus is in the microwave region. The transition between the $|11-1\rangle$ state and $|210\rangle$ state is about 31 MHz far off resonance. The transition process of a two-level system is well known as Rabi oscillation,[18] and the Rabi frequency of our σ^+ circularly polarized microwave molecular rotational level transition is

$$\Omega = -\frac{E_0}{\sqrt{2}\hbar}\langle J+1, K, M+1|\mu_X + i\mu_Y|J, K, M\rangle, \tag{5}$$

where E_0 is the amplitude of the microwave electric field, $\hbar$ is the reduced Planck constant, μ_X and μ_Y are the components of molecular dipole moment μ along the space-fixed X axis and Y axis, respectively. The dipole moment matrix elements of μ_X and μ_Y can be found in textbook,[16] and we obtain

$$\Omega = \frac{E_0\mu}{\sqrt{2}\hbar}\frac{[(J+1)^2-K^2]^{1/2}[(J+M+1)(J+M+2)]^{1/2}}{(J+1)[(2J+1)(2J+3)]^{1/2}}. \tag{6}$$

For the transition between the states $|1-11\rangle$ and $|2-12\rangle$, $\Omega = \sqrt{3/10}E_0\mu/\hbar$.

Now the molecules are composed of rotational states $|11-1\rangle$ and $|2-12\rangle$. If the detecting light only interacts with the $|2-12\rangle$ state, the gravitational acceleration g of molecules with an upward orientation of angular momentum can be measured.

To measure the free fall acceleration g of molecules in a certain rotation state, a Doppler sensitive laser beam can be used to excite molecules from a specific rotational level in the ground vibrational state into a specific rotational level in an excited vibrational state,

such as a CO_2 laser of 9.494 μm which can excite the $v_3 = 0 \to 1$, $J = 2 \to 3$ transition of the CH_3F molecule. The v_3 vibrational state corresponds to the C-F stretching vibration. Molecules in $J = 1$ rotational state have no transition line near-resonant, thus can not be detected. As molecules free fall, they have an overall Doppler shift relative to the laser which propagates vertically. If we know the Doppler shifts $\Delta\omega_1$ and $\Delta\omega_2$ at times t_1 and t_2, we can obtain the g value as

$$g = \left| \frac{\Delta\omega_2 - \Delta\omega_1}{k(t_2 - t_1)} \right|, \tag{7}$$

where k is the wave vector of the laser, and $t_2 - t_1$ is the time interval of measurements which can be assumed to be 0.2 s for about 20 cm free fall. The relative uncertainty of the frequency measurements can be estimated by the Allan standard deviation[19] as $\sigma(\tau) = 1/(Q \cdot \mathrm{SNR} \cdot \sqrt{\tau})$, where Q is the line quality factor (transition frequency/FWHM), SNR is the signal-to-noise ratio, and τ is the total measurement integral time. The FWHM of the line shape have three compositions: natural line width, Doppler broadening and collision broadening. The natural line width (about 50 Hz) and the collision broadening (less than 1 Hz) are neglectable in comparison with the Doppler broadening, which is about 2.4 MHz at the temperature of 10 mK. The SNR due to the shot-noise limit can be written as $\sqrt{N}$, where N is the number of detected molecules. If N is about 10^5 at t_1, then due to the thermal diffusion of molecules, it is about 10^3 at t_2. The total measurement time τ is assumed to be 100 s, and we can finally deterimine the uncertainty of the frequency measurement, which is 0.77 kHz at time t_1 and 7.7 kHz at time t_2, corresponding to the uncertainty of the g measurement at the level of $10^{-3}g$ at 100 s integral measurement time.

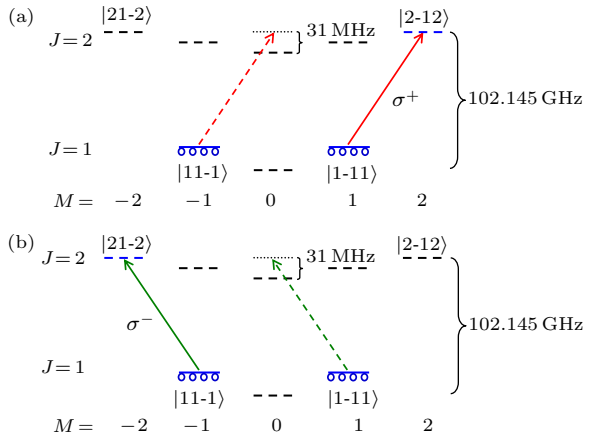

Fig. 2. Transition between the $J = 1$ and the $J = 2$ levels driven by a circularly polarized microwave field. Molecules are initially populated in the states $|11-1\rangle$ and $|1-11\rangle$. A bias static electrical field is applied to split different M levels. (a) For the σ^+ polarized field, states $|1-11\rangle$ and $|2-12\rangle$ are coupled, and (b) for the σ^- polarized field, states $|11-1\rangle$ and $|21-2\rangle$ are coupled.

In order to measure the g of molecules with a downward orientation of angular momentum, we should use a σ^- circularly polarized microwave to transfer the $|11-1\rangle$ state to the $|21-2\rangle$ state, as shown in Fig. 2(b), and detect the $|21-2\rangle$ state only.

The above discussion is based on the condition of molecular translational temperature at the mK level; if we can further cool down the molecules to the μK level, the gravitational acceleration of molecules can be measured by taking advantage of the phenomenon of the Bloch oscillations occurring to particles driven by a constant force in periodic potential, or Raman-type matter-wave interferometers.[20] Recently, some experiments applying Bloch oscillations of cold atoms to gravity measurement have been reported,[21] and the gravitational acceleration g can be measured at 10^{-7} relative precision. For this method of g measurement, we should let the molecules already manipulated by the microwave field fly again through the hexapole field to remove the molecules in undesirable rotational states. In the proposed experiment, polarized rotational state molecules are trapped in a one-dimensional vertical optical lattice created by two interfering laser beams and the periodic potential results from the ac-Stark shift of molecules illuminated by a laser standing wave. The molecules under gravity execute Bloch oscillations at a frequency ν_{B} given by

$$\nu_{\mathrm{B}} = \frac{mg\lambda_L}{2h}, \tag{8}$$

where λ_L is the wavelength of the light producing the optical lattice, and h is the Planck constant. The Bloch frequency ν_{B} can be measured by a time-of-flight scheme after letting the molecules evolve in the optical lattice or monitoring the *insitu* wave-packet extent. The differential free fall acceleration Δg between different polarized rotational state molecules can be obtained by comparing the Bloch frequency difference $\Delta\nu_{\mathrm{B}}$ as follows:

$$\Delta g/g = \Delta\nu_{\mathrm{B}}/\nu_{\mathrm{B}}. \tag{9}$$

An assumed 10^{-7} relative precision of the measurement of the Bloch frequency ν_{B} of molecules will enable us to test the violation of the equivalence principle $\Delta g/g$ at the same level.

In the Raman-type matter-wave interferometer, atoms free fall in the gravitational field, and two counter-propagating laser beams, whose wavevectors are designated as $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$, stimulate Raman transitions between two hyperfine ground states of the atoms. The g value is included in the expression of the interferometer phase,

$$\varphi_g = \boldsymbol{k}_{\mathrm{eff}} \cdot \boldsymbol{g}T^2, \tag{10}$$

where $\boldsymbol{k}_{\mathrm{eff}} = \boldsymbol{k}_1 - \boldsymbol{k}_2$ is the effective wavevector, and T is the time interval of the optical pulses. The differential free fall acceleration Δg can be obtained as follows:

$$\Delta g/g = \Delta\varphi/\varphi_g, \tag{11}$$

where $\Delta\varphi$ is the interferometer phase difference between different polarized rotational state molecules. If the Raman transitions occur between suitable quantum states in molecules, e.g. the molecular rotational

states $|1-11\rangle$ and $|2-12\rangle$, the gravitational acceleration g can be measured using the similar method, and the measurement precision has the potential to reach the level of $10^{-9}g$. We have performed a series of atom interferometer experiments in gravity measurement, and the systematic errors are under analyzed.[22–24]

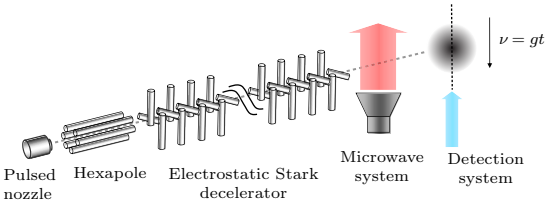

Fig. 3. The schematic diagram of the experimental apparatus.

To test the equivalence principle with rotating cold polar molecules, we should polarize their rotational states and compare the gravitational acceleration difference Δg of them. As discussed above, to increase the experimental precision, molecules should be further cooled down. Due to the complicated level structure, the laser cooling, which is widely applied to atoms, is essentially inapplicable to molecules. However, some different techniques,[25] such as photoassociation[26] or Feschbach resonances,[27] buffer-gas cooling,[28] electrostatic Stark deceleration,[29] and optical Stark deceleration,[30] have developed to generate slow and cold molecules. Among these methods, we prefer using the technique of electrostatic Stark deceleration, which can not only vary the velocity of polar molecules, but also keep the rotational state of the molecules unchanged during the process of deceleration. The Stark decelerator takes advantage of the molecular Stark effect to decelerate and cool down molecules and molecules can be decelerated to a standstill.

Incidentally, atoms can have electron orbital angular momentum or nuclear "spin" which typically has substantial contributions from orbital angular momentum, and the experimental methods for cooling, trapping, and detection are more advanced. Thus we can perform similar experiments with atoms.

In summary, we have proposed a novel scheme for testing the equivalence principle with rotating cold molecules whose angular momentum are polarized vertically up and down, and the proposed experimental setup is illustrated in Fig. 3. We use the hexapole state-selection technique to select molecules in specific rotational states and cool them down using the electrostatic Stark deceleration technique to the mK level. A microwave field is used to manipulate the rotational states of molecules to ensure that molecules with particular spatial angular momentum orientation are detected during the process of the g measurement. The relative precision of the g measurement is estimated to be at the level of 10^{-3}. If the translational temperature of the molecules can be further cooled down to the μK level, it is hopeful that advantage can be taken of the phenomenon of the Bloch oscillations to compare the g at the level of 10^{-7}, or even use a Raman-type matter-wave interferometer to compare the g at the level of 10^{-9}. Of course, some fundamental physical limitations of the measurement of the gravitational acceleration g of molecules should be concerned, such as Coriolis force, inhomogeneous electric and magnetic field, background vibration and dipole interaction between molecules. An observed difference between the gravitational accelerations of two kinds of molecules would imply violation of the equivalence principle and would help to explore the Spatio-temporal topological structure. Of course, there are many challenges in this proposed experiment, and we will try to realize it step by step.

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