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# 3 Atomic Frequency Standards

## 3-1 Basic Physics in the Atomic Frequency Standards

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Today, the second, unit of time, is defined by using the hyper fine transition of cesium atoms. In atomic clocks and atomic frequency standards, various quantum transitions are used. To understand how these equipments works, elementary and intuitive illustrations are show in this paper. The main topics are the energy level of atoms, life time, quantum transition and radiation, and the Ramsey resonance for the precise measurement of the transition frequency.

### *Keywords*

Atomic frequency standards, Energy level, Quantum transition, Life time, Ramsey resonance

### 1 Introduction

As discussed in Article **2-1** of this special issue[1], the second is currently defined based on the period of the radiation corresponding to the transition between the hyperfine structure levels of the cesium atom. Although current science has by no means elucidated the full range of natural phenomena, the description of the energy structures of atoms by quantum mechanics is based on principles and constants that are universal and time-invariant, and thus this description is extremely accurate. It is now widely accepted that radiations corresponding to the quantum transitions of atoms are most suitable for defining and realizing time and frequency standards.

To understand the definition and the mechanism of atomic frequency standards, a broad base of knowledge is indispensable, encompassing electromagnetics, quantum mechanics, and atomic physics. A standard reference work is available[2] with a detailed explanation of the subject, but it is extremely voluminous and hard to read through. Other valuable publications[3][4] outline the subject more sim-

ply, including textbooks for time and frequency standards. Here I will attempt to explain the relevant basic physics as intuitively as possible, to provide a base of knowledge for those approaching these textbooks. The major topics I will discuss include atomic energy levels, quantum transitions (such as the electric dipole transition), and Ramsey resonance. A knowledge of mechanics, electromagnetics, and quantum mechanics equivalent to that of a second or third-year student at a science or technological college should be sufficient to understand the following descriptions.

As I obviously cannot explain every relevant point in this short article, please see references[2],[3],and[4] for further details. I hope that this article will provide the reader with the basic concepts required to understand these texts.

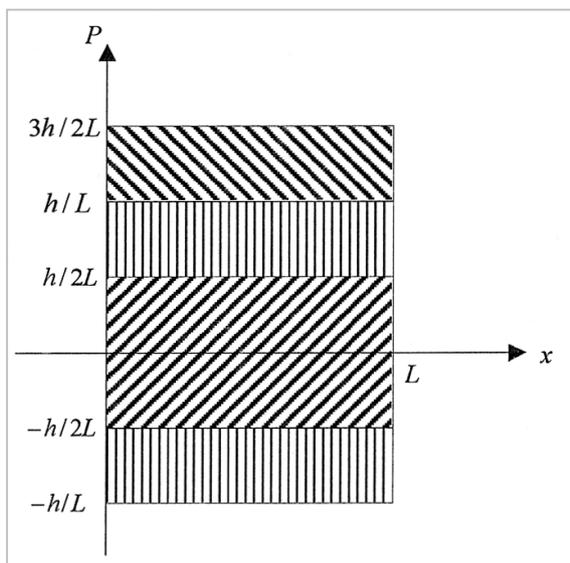
### 2 Atomic spectra and energy levels

The characteristics of quantum theory can be roughly stated as follows: phase space (the virtual space whose axes are the positions and

momentums of the particles that constitute the system) is discretely divided into a number of regions, each of which has an area of the size of the Planck constant,  $h$ , and takes a physical state that cannot be reduced any further. As a result, it can be shown that discrete energy states (energy levels) are present within a bound system. In understanding this effect, the description of the harmonic oscillator in the quantum mechanics textbook by Tomonaga[5] serves as an excellent reference. However, a similar effect can also be observed in the simpler and more extreme example of a square-well potential.

Let us consider a square-well potential in which the potential is zero in a region of length  $L$ , given an infinitely large exterior region. The particles contained in the well diffuse in the region between 0 and  $L$ . Let us assume that the phase space is divided into discrete divisions which have the Planck constant  $h$ . The spatial range is confined within  $L$ , and in the wave description, the forward and reverse waves are superposed to form a standing wave. Thus, the momentum  $P_n$  of the  $n$ -th state is considered to take a discrete value, as follows (Fig.1):

$$P_n = \frac{h}{2L}n, \quad n=1,2,3\cdots \quad (1)$$



**Fig. 1** Phase space of the square-well potential system and the quantized region

Thus, denoting the mass of the particle by  $m$ , the energy,  $E_n$ , of the  $n$ -th state is obtained as

$$E_n = \frac{P_n^2}{2m} = \frac{h^2 n^2}{8mL^2} \quad (2)$$

without solving the Schrödinger equation. In this way, in accordance with quantum theory, momentum and energy level take discrete values in a system confined to a finite region.

Photons, quanta of electromagnetic waves, are considered to propagate infinitely once generated if not confined within a finite region. The energy in this case takes continuous values and satisfies the well-known Einstein formula for light quanta:

$$E = h\nu \quad (3)$$

Viewing this equation together with the formulae  $c = \lambda\nu$  for the wave, and  $E/c = p$  for the photon, we may also conclude that the product of the photon wavelength and momentum is quantized with the Planck constant. Further, angular momentum is an important physical quantity in atomic physics. It can be regarded as the generalized momentum conjugate to the angle, which only takes values from 0 to  $2\pi$  radians, so that the maximum range of uncertainty is  $2\pi$ . For this reason, the angular momentum is quantized to the parts whose values are  $h/2\pi$ . In this manner, the region in the phase space with a finite area of the size of the Planck constant  $h$  takes a quantum state. This differs fundamentally from the classical idea that each point in phase space represents an independent physical state. However, this difference aside, we must not forget that the descriptions of phenomena provided by classical physics are also effective in most cases.

With regard to the finite or infinite nature of the system, an atom is a finite bound state in which electromagnetic interactions bind the electrons in a region of approximately 0.1 nm around the small nucleus. Thus, the physical system of the atom has discrete energy levels. Considering that the electrons are bound in a region of approximately  $10^{-10}$  m by the poten-

tial generated by the nucleus, it is roughly described that the electrons bound in a well potential of the size of the atom make the energy level. We can estimate the energy levels in an atom by calculating the energy levels of an electron in the square well potential. Denoting the Planck constant as  $h$ , the mass of the electron as  $m$ , and the region in which the electron is confined as  $L$ , Eq. (2) gives the following expression for the energy levels of the confined electron:

$$E_n = \frac{h^2 n^2}{8mL^2} \quad (4)$$

Given a Planck constant  $h$  of  $6.63 \times 10^{-34}$  [J·s], an electron mass  $m$  of  $9.11 \times 10^{-31}$  [kg], and length  $L$  of  $1.0 \times 10^{-10}$  [m], the energy value for  $n = 1$  is

$$E_{n=1} = \frac{(6.63 \times 10^{-34})^2 [\text{J} \cdot \text{s}]^2}{8 \times 9.11 \times 10^{-31} [\text{kg}] \times (1 \times 10^{-10})^2 [\text{m}]^2} \quad (5)$$

$$= 6.04 \times 10^{-18} [\text{J}]$$

The frequency  $\nu$  of the photon corresponding to this energy is obtained from  $E = h\nu$ :

$$\nu = \frac{6.04 \times 10^{-18} [\text{J}]}{6.63 \times 10^{-34} [\text{J} \cdot \text{s}]} \quad (6)$$

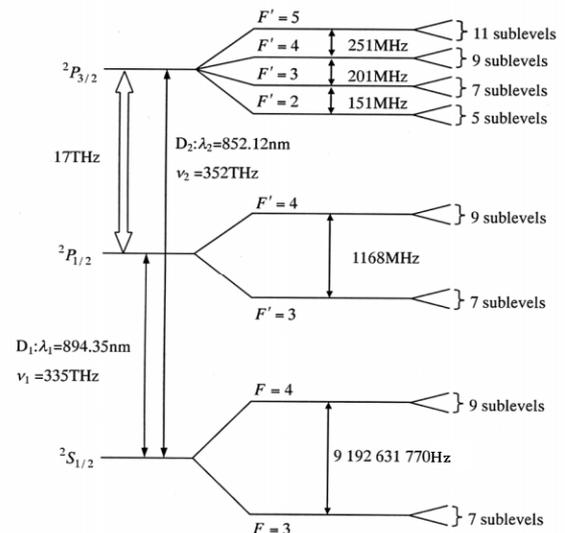
$$= 9.10 \times 10^{15} [\text{Hz}]$$

This energy is the value needed to excite a particle in the zero kinetic energy state in a square well potential to the next higher level, but is also considered to be of approximately the same order of magnitude as the energy required to release a particle bound in a finite potential. Of course, the details of the value largely depend on the shape of the potential, but the value here corresponds to the short ultraviolet wavelength, which agrees with the finding that the ultraviolet range is required for the ionization of atoms. A square well potential is far from a realistic model for the atom, but it is effective in estimating the order of magnitude of the relevant phenomena.

The energy levels of an atom are more precisely calculated based on the example of the hydrogen atom, where electrons are bound in a Coulomb potential described in many textbooks [6]-[8]. The levels depend not only

on the principal quantum number and the orbital quantum number, which both appear in the expression for the hydrogen atom, but also on other internal and external factors such as orbital angular momentum, spin, the interactions between these factors, and the external electromagnetic field, all of which complicate the problem. Here it is common practice to use the following notations: orbital angular momentum for the electron is  $l$ , its spin is  $s$ , and its total angular momentum, which is the sum of these two, is  $J$ . The states with  $l = 0, 1, 2 \dots$  are labeled the  $S$  state, the  $P$  state, the  $D$  state, and so on, respectively. Nuclear spin is denoted as  $I$ . The quantity obtained by combining all the angular momentums of the electron and the nucleus is called  $F$ .

When energy levels are close to each other, these levels are referred to as fine structures. These structures are usually formed due to spin-orbit interaction. When energy levels are even closer together due to interactions such as spin-spin magnetic interactions, these levels are called hyperfine structures. Let us consider these in the example of the cesium atom (Fig.2).



**Fig.2** Energy levels of the cesium atom: fine and hyperfine structures

The frequency corresponding to the transition between the  $S$  state, which is the ground state, and the  $P$  states, which is the first excited state of the angular momentum, is over 300

THz. The  $P$  states are divided into two states corresponding to orbital-spin combined angular momentums with values of  $1/2$  and  $3/2$ , respectively, with a resultant frequency difference of 17 THz. This is a fine structure. A cesium nucleus has a nuclear spin of  $I = 7/2$ . In the ground state of the atom,  $l = 0$  holds and the total angular momentum  $F$  (combining the nuclear spin of  $7/2$  and the electron spin of  $1/2$ ) is either 3 or 4, which results in a small difference in the energy levels. This is a hyperfine structure, and the frequency corresponding to the transition between these energy levels is approximately 9 GHz. It is clear that the intervals of the fine structures are only several percent of those of normal energy levels and that the hyperfine structures exhibit intervals approximately one thousandth the size of those of fine structures. In the magnet field, each level splits further into sublevels, due to the Zeeman effect, according to the total angular momentum (including nuclear angular momentum).

### 3 Interaction between atoms and electromagnetic waves

If a quantum state is the energy eigenstate in a strict sense, once the state is established, it will continue to exist infinitely, without any transitions to other states. However, in reality, most quantum states undergo transitions to other states in finite time through various interactions. In particular, when the atom is excited to a higher-energy state, it will undergo transitions to lower-energy states spontaneously through intra-atomic interactions even without any energy provided from outside the atom. If the excited state undergoes a transition in this way (through a given interaction) to a lower-energy level with a certain probability per unit time, the probability that the atom will stay in the initial state steadily decreases with time. When this is the case, the excited energy level is said to have a finite lifetime. Here "lifetime" is most often defined as the time after which the probability that the system will continue to remain in the initial

state becomes  $1/e$ . The time after which the probability of existence becomes  $1/2$  is referred to as the well-known "half-life."

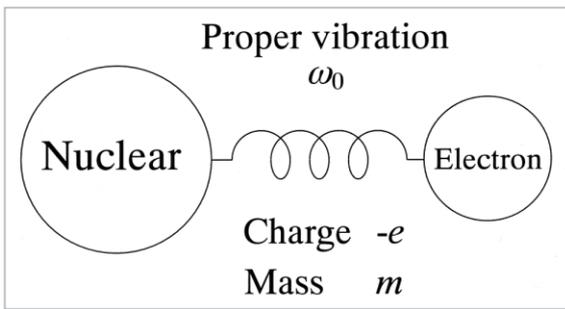
To say that an energy level has a lifetime is to say that the level no longer has a definite energy value but that it does have a finite width. In this sense, the frequency of the quantum transition between two energy levels also has a finite width instead of a definite value. When applying the quantum transition to frequency standards, it is desirable to use a transition that features a narrow frequency width and occurs between states with lifetimes that are as long as possible. Although transitions do not readily occur from states with long lifetimes, there are methods (such as induced emission) that applies external forces to effect a transition[9]-[11]; such control thus induces transitions with the desired long lifetimes.

To calculate the lifetime of an excited state precisely, a proper quantum mechanical treatment is required. However, for an intuitive understanding of the concept, a fully quantum treatment is not necessary; instead, a semi-classical treatment can provide an approximate idea. Using the classical theory may give an incorrect description of the elementary processes, but the resultant macroscopic quantities are meaningful. This is also expected from Ehrenfest's theorem, which states that the motion of the expected value of the wave function equals classical motion when the potential change is negligible within the extent of the wave packet. Thus, when treating a large number of atoms, the semi-classical descriptions of quantities are sufficiently meaningful, as quantities corresponding to the sum of random quantum processes. In addition, this treatment is valuable in the sense that the partial, precision application of quantum theory alongside the classical theory provides a relatively concrete understanding of the concepts involved, greatly contributing to an intuitive understanding of the relevant phenomena and enabling estimation at the required order of magnitude. However, it should be noted clearly that the model used here is applied in

order to understand certain aspects of the phenomena, and that it is used metaphorically, with many features that are inconsistent with reality.

Here, we will treat the problem based on the classical theory and use the quantum theory partially, to describe the quantum transition within the atom, as well as to explain the phenomena numerically[12].

First, in relation to the basic electric dipole transition, let us consider the classical model of simple harmonic oscillation of a dipole charge (Fig.3).



**Fig.3** Classical model for electric dipole radiation of an atom

Let us assume that an electron with charge  $-e$  and mass  $m$  is bound to a heavy nucleus with spring constant  $m\omega_0^2$ , (eigen frequency being  $\omega_0$ ) and that it is damped weakly, at a rate of  $1/\tau$  by dipole radiation as it oscillates. Assuming that the motion of the nucleus is negligible, the energy loss of the atom is due only to the motion of the electron. Further, as the damping is extremely weak, this damped oscillation and its energy value can be approximated using simple harmonic oscillation at each instant. In other words, let us assume that the energy  $E(t)$  of the atom is approximately expressed as

$$E(t) = \frac{1}{2} m \omega_0^2 x^2(t) + \frac{1}{2} m \dot{x}^2(t) \quad (7)$$

but is gradually damped to

$$E(t) = E_0 e^{-\frac{t}{\tau}} \quad (8)$$

during which the following holds:

$$\omega_0 \gg 1/\tau \quad (9)$$

Denoting the dipole moment as  $\mathbf{d}$ , the power radiated by this oscillation is expressed as follows[13]:

$$P = \frac{2}{3} \frac{\ddot{\mathbf{d}}^2}{c^3} \quad (10)$$

Here, let us consider the dipole to consist of simple harmonic oscillation of a charge with an amplitude about the size the atom itself. Denoting the size of the atom as  $a$ , the position of the charge  $x$  in the one-dimensional direction of the oscillation is expressed as follows:

$$x = a \sin \omega_0 t \quad (11)$$

and from

$$\mathbf{d} = ex = ea \sin \omega_0 t \quad (12)$$

the following is obtained:

$$\ddot{\mathbf{d}} = -e\omega_0^2 x \quad (13)$$

Inserting this into Eq. (10) gives a power value of:

$$P = \frac{2}{3} \frac{e^2 \omega_0^4 x^2}{c^3} \quad (14)$$

Ignoring the details of the coefficients and assuming that the average of  $x$  is roughly in the same order of magnitude as oscillation amplitude  $a$ , the average of radiated power  $\langle P \rangle$  is

$$\langle P \rangle \sim \frac{e^2 \omega_0^4 a^2}{c^3} \quad (15)$$

Here, the radiated energy over  $\tau$  seconds is estimated to be approximately  $\langle P \rangle \tau$ . Assuming that this is approximately equal to the energy of the light quantum with angular frequency  $\omega_0$  radiated through the elementary process of the quantum transition, and using  $E_0 \sim \langle P \rangle \tau \sim \hbar \omega_0$  and Eq. (15),

$$\tau \sim \frac{\hbar c^3}{e^2 \omega_0^3 a^2} \quad (16)$$

is obtained. Applying the fine structure constant  $\alpha = e^2/\hbar c$  to this gives the following expression:

$$\tau \sim \frac{1}{\alpha} \left( \frac{c}{\omega_0 a} \right)^2 \frac{1}{\omega_0} \quad (17)$$

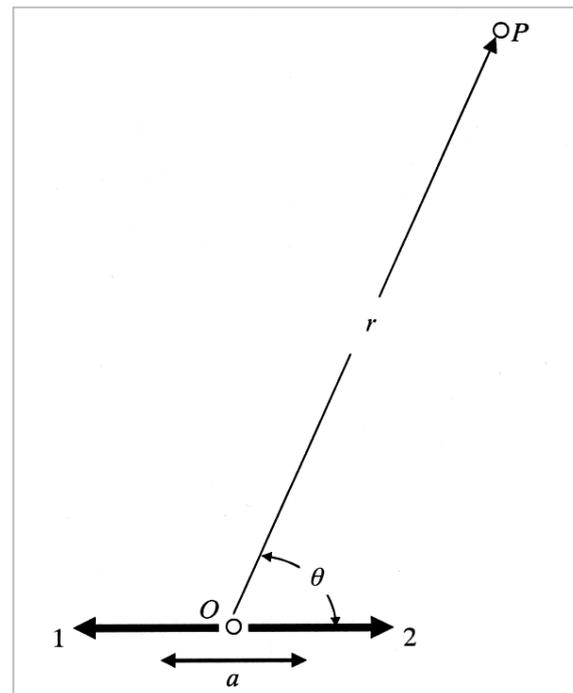
Here, the oscillation amplitude  $a$  of the dipole is assumed to be approximately the size of the atom,  $10^{-10}$  m. It should also be noted that  $(c/\omega_0 a)$  approximately equals the wavelength  $\lambda$  of the radiation. The lifetime of the excited state and the oscillation period of the radiation upon decay (two values sharing the same dimension for a single phenomenon) are of largely different orders of magnitude. From these results, we can see the reasons of the large difference as follows: in the excited state, the fine structure constant is much smaller than 1 and the wavelength of the radiation is much larger than the size of the atom; thus the lifetime becomes larger than the oscillation period ( $1/\omega$ ) of the radiated electromagnetic wave by a factor equal to the product of the fine structure constant and the square of the ratio of wavelength to atomic size, as indicated in Eq. (17). Observing only lifetime and radiation frequency, it is clear that the excitation lifetime is inversely proportional to the cube of the radiation frequency of the quantum transition.

When the radiation is visible light ( $\omega \sim 3 \times 10^{15}$ ,  $\lambda \sim 600$  nm), it is understood from  $\tau \sim 137 \times 600^2 / 3 \times 10^{15} \sim 2 \times 10^{-8}$  s that lifetime is approximately several tens of nano-seconds if the transition is due to electric dipole radiation of atomic size oscillation. This is in fact a good approximation for the excited states of many atoms. However, a quantum transition with a much longer lifetime (known as the "forbidden transition") cannot be understood with this explanation. If this explanation is valid in the microwave regions, a long lifetime is expected due to low frequency. There are, however, some other reasons for the long lifetime of the transition of the microwave standards.

The transition of the hyperfine structure, used in the definition of time and frequency standards, is a magnetic dipole transition and features an even longer lifetime than the electric dipole transition. Further, the forbidden

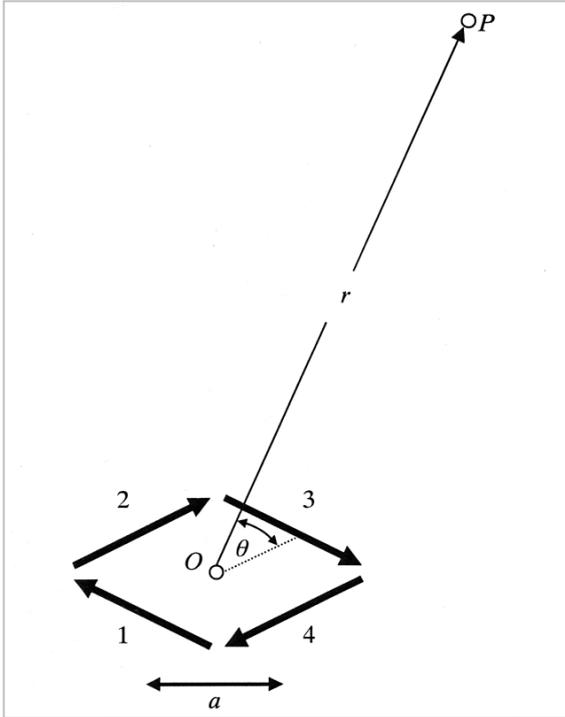
transition is another transition that does not involve the electric dipole, due to the symmetry of the atomic structure; instead, the transition is caused by electric quadrupoles or magnetic dipoles. The classical view of the longer lifetimes of these excited states (relative to the electric dipole) can be described in the following manner.

Electric quadrupoles and magnetic dipoles can be constructed with electric dipoles. As shown in Fig.4, when two electric dipoles oscillating in opposite directions are placed on the same axis at a distance apart of  $a$ , corresponding to the size of the atom, the electric dipole oscillations cancel each other out and electric quadrupole oscillation is generated. As shown in Fig.5, if four electric dipoles are placed on the sides of a square, the electric dipole oscillations and the electric quadrupole oscillations all cancel out and an oscillating current in the form of a ring is generated; this is magnetic dipole oscillation.



**Fig.4** Electric quadrupole oscillation formed by two electric dipole oscillations

In these cases, the electromagnetic fields of the paired electric dipoles do not completely cancel each other out; instead, a weak electromagnetic field remains (except on the spe-



**Fig.5** Magnetic dipole oscillation formed by four electric dipole oscillations

cial axis equidistant from the respective dipoles). For example, in Fig.4, at the position on the  $x$ -axis at a distance  $r$  from the center of the dipole pair, the electric field generated by the nearer of the two electric dipoles parallel to the  $y$ -axis is assumed to be

$$E_1 = A(r - a/2)e^{i\omega\left(t - \frac{r-a/2}{c}\right)} \quad (18)$$

The electric field generated by the far side electric dipole shows anti-phase oscillation:

$$E_2 = -A(r + a/2)e^{i\omega\left(t - \frac{r+a/2}{c}\right)} \quad (19)$$

The difference of  $\pm a/2$  in amplitude  $A(r)$  could be neglected at a sufficiently distant position, but the phase difference remains. Superposing these two electric fields gives the resultant electric field:

$$E_2 \sim i \frac{a\omega}{c} E_1, \quad (20)$$

This is similar to the case of magnetic dipole oscillation, in which the electromagnetic fields generated by opposite paired electric dipoles cancel each other out. In other words,

for electric quadrupole oscillation or magnetic dipole oscillation, the amount of electric field cancelled by the paired electric dipoles increases as the wavelength of the radiated electromagnetic wave becomes larger relative to the size of the atom. The resultant electric field in this case becomes as small as approximately  $\left(\frac{a\omega}{c}\right)$  times the size of the field generat-

ed by the electric dipoles. As radiated energy is proportional to  $E^2$ , this energy in electric quadrupole oscillation or magnetic dipole oscillation becomes approximately  $\left(\frac{a\omega}{c}\right)^2$  times

smaller than with electric dipole oscillation, and the lifetime becomes correspondingly longer. The extremely long lifetime of the forbidden transition resulting from electric quadrupoles or magnetic dipoles when the electric dipole transition is forbidden due to the symmetry of the atom can be understood in light of these conclusions. In the region of

visible light, the applicable ratio is  $\left(\frac{a\omega}{c}\right) \sim 10^{-3}$ ;

thus this effect lengthens the lifetime by 6 to 7 orders of magnitude, which brings the lifetime of excitation by the forbidden transition up to

nearly 1 second. This ratio is  $\left(\frac{a\omega}{c}\right) \sim 10^{-7}$  for

microwaves, extending the magnetic dipole transition lifetime by 14 to 15 orders of magnitude. The results of these semi-classical analyses show that the lifetime of the magnetic dipole transition between hyperfine structures in the microwave region will exceed the age of the universe ( $\sim 10^{17}$  seconds) if no external perturbation is applied.

#### 4 Cs atomic spectrum and Ramsey resonance

The frequency of the radiation corresponding to the transition of the hyperfine structure of the cesium atom, used in the current defini-

tion of the second, is measured using the phenomenon known as Ramsey resonance for many atomic clocks and for all of the primary frequency standards. Applying Ramsey resonance is not only an important technique in cesium atom applications but is also indispensable in the precise measurement of the frequencies of quantum transitions.

Ramsey resonance is induced as follows: create a constant magnetic field, known as the C magnetic field, in order to split the degeneration of energy levels, thus forcing the atoms to take a certain state. Irradiate the atoms with an electromagnetic wave in this state for a period of time  $\tau$ , wait for time  $T$ , and irradiate the atoms with the electromagnetic wave again for time  $\tau$ . When allowing the atoms to interact with the electromagnetic wave in two separate processes in this manner, the probability that an atom will undergo a transition to another state depends not only on the interaction time  $\tau$ , but also on the elapsed time between the repeated interaction processes. A term resembling the interference term then appears in the transition probability, and it can be demonstrated that the quantum transition becomes extremely sensitive to frequency change. This is Ramsey resonance. Using this phenomenon, the central frequency of the transition can be measured with high precision.

Before performing the slightly complicated calculations involved, I will state the results here beforehand, to clarify the procedure. The probability  $P$  that the atom has undergone a quantum transition after the Ramsey resonance processes is expressed as follows:

$$P(\tau) = \frac{1}{2} \sin^2 b \tau [1 + \cos(\omega - \omega_0)T] \quad (21)$$

Here,  $\omega$  is the frequency of the irradiated electromagnetic wave,  $\omega_0$  is the central frequency of the radiation emitted by the transition (which can be calculated from the difference between the energy levels, and  $b$  is a quantity determined by the intensity of the irradiated electromagnetic wave, which can be

expressed using the Bohr magneton  $\mu_B$ , and magnetic flux density  $B$ :

$$b = \frac{\mu_B B}{\hbar} \quad (22)$$

The full width at half maximum of the frequency of the transition probability  $P$  given by Eq. (21) is

$$\Delta\nu_0 = \frac{1}{2T} \quad (23)$$

In this way, taking large values for the time interval between the two interaction processes induced by the irradiation of the electromagnetic wave can significantly narrow the frequency range in which the transition occurs. However, it should also be noted that Eq. (21) is an extremely simplified expression and is valid only when the condition

$$|\omega - \omega_0| \ll b \quad (24)$$

is satisfied. For a thermal beam standard in which the atoms are ejected and the electromagnetic wave is irradiated at different positions, this condition is also expressed as follows:

$$\frac{|\nu - \nu_0|}{\Delta\nu_0} \ll \frac{L}{l} \frac{b \tau}{\pi} \quad (25)$$

Here,  $l$  is the length of the respective irradiation region,  $L$  is the distance between the two irradiation regions, and the relation  $T/\tau = L/l$  and Eq. (23) have been applied. We are only considering cases in which this simplifying condition holds.

There are several methods for deriving the probability of Ramsey resonance. One well-known method uses the density matrix[2], but a more direct—and in a sense clearer—method is presented below, involving solution of the Schrödinger equation[4].

The hyperfine structures can be approximately expressed in terms of a two-level model in which a weak interaction exerts a force on the energy eigenstates. Particularly for the clock transition of cesium 133, if the appropriate C magnetic field is imposed and the direction of the interacting electromagnetic

wave is correct, it can be assumed that only the transition between the ground state  $F=4$ ,  $m_F=0$  and the state  $F=3$ ,  $m_F=0$  applies, and that no other transition occurs.

Here, denoting the state  $F=4$ ,  $m_F=0$  as  $|\Psi_1\rangle$  and the state  $F=3$ ,  $m_F=0$  as  $|\Psi_2\rangle$ , the general quantum state corresponds to the superposition of the two states, which can be expressed by the following state function:

$$|\psi\rangle = c_1|\Psi_1\rangle + c_2|\Psi_2\rangle \quad (26)$$

Here,  $c_1$  and  $c_2$  are the normalized coefficients for the superposition. Limiting the number of states to two can simplify the phase space to two dimensions, and the states can be expressed as follows:

$$|\Psi_1\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad |\Psi_2\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \quad (27)$$

The unperturbed Hamiltonian diagonalized in this two-dimensional phase space for the states  $F=4$ ,  $m_F=0$  and  $F=3$ ,  $m_F=0$  is expressed as follows:

$$H_0 = \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & 0 \\ 0 & \omega_0 \end{pmatrix} \quad (28)$$

On the other hand, the non-diagonal Hamiltonian perturbation for the interaction with the microwave is expressed as follows:

$$H_1 = \frac{\hbar}{2} \begin{pmatrix} 0 & 2b \cos \omega t \\ 2b \cos \omega t & 0 \end{pmatrix} \quad (29)$$

Added together, these two expressions constitute the Hamiltonian  $H$  for an atom interacting with the electromagnetic wave. The Schrödinger equation using this Hamiltonian is expressed as follows:

$$i\hbar \frac{d}{dt} |\psi\rangle = H |\psi\rangle \quad (30)$$

Substituting the state function described above into Eq. (30) and expanding it yields the following equations for the superposition coefficients:

$$i \frac{dc_1}{dt} = -\frac{\omega_0}{2} c_1 + \frac{b}{2} [\exp(i\omega t) + \exp(-i\omega t)] c_2 \quad (31)$$

$$i \frac{dc_2}{dt} = \frac{b}{2} [\exp(-i\omega t) + \exp(i\omega t)] c_1 + \frac{\omega_0}{2} c_2 \quad (32)$$

If these equations can be solved to obtain the time dependencies of  $c_1$  and  $c_2$ , the transition probability can be calculated. It should be noted when solving these equations that the elapsed time for the atom in the traditional sense (related to  $H_0$ ) should be distinguished from the elapsed time for the interaction with the electromagnetic wave (related to  $H_1$ ). Denoting the former by  $t$  and the latter by  $\theta$ , the time differential applicable to this problem can be expressed as follows:

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \frac{\partial}{\partial \theta} \quad (33)$$

Accordingly, the time dependencies of  $c_1$  and  $c_2$  can also be separated into two parts: one part depending on  $t$  and one part depending on  $\theta$ . Particularly in this case, parts depending on  $t$  are considered to correspond to energy eigenstates, which are expressed as  $\exp(\pm i\omega t/2)$ , and these are multiplied by the perturbation terms, which are functions of  $\theta$ . Denoting these perturbation terms as  $\gamma_1$  and  $\gamma_2$ ,  $c_1$  and  $c_2$  can be rewritten as follows:

$$c_1(t, \theta) = \gamma_1(\theta) \exp \frac{i\omega t}{2} \quad (34)$$

$$c_2(t, \theta) = \gamma_2(\theta) \exp \left( -\frac{i\omega t}{2} \right) \quad (35)$$

Substituting these into Eqs. (31) and (32) gives the following equations for  $\gamma_1$  and  $\gamma_2$ :

$$i \frac{\partial \gamma_1}{\partial \theta} = \frac{\Omega_0}{2} \gamma_1 + \frac{b}{2} \gamma_2 \quad (36)$$

$$i \frac{\partial \gamma_2}{\partial \theta} = \frac{b}{2} \gamma_1 - \frac{\Omega_0}{2} \gamma_2 \quad (37)$$

Here  $\Omega_0$  is the quantity defined as follows:

$$\Omega_0 = \omega - \omega_0 \quad (38)$$

Eqs.(36) and (37) are approximated expressions obtained neglecting the terms depending on  $2\omega$ . In a more strict treatment, this term will give rise to a high-frequency term of  $2\omega$  in the solution, but it is understood that in such treatment the amplitude is negligibly small. Treating this term as of negligible

significance simplifies the calculation as illustrated below.

As shown in Eqs.(34) and (35),  $\gamma_1$  and  $\gamma_2$  express the states of the system. The time evolution of this system during  $\theta$  is formally expressed in the matrix expression as follows:

$$\begin{pmatrix} \gamma_1(\theta + \theta_0) \\ \gamma_2(\theta + \theta_0) \end{pmatrix} = M^{(k)}(\theta) \begin{pmatrix} \gamma_1(\theta_0) \\ \gamma_2(\theta_0) \end{pmatrix} \quad (39)$$

Here, the matrix  $M^{(k)}(\theta)$  is obtained as the solution of Eqs.(36) and (37). The superscript  $k$  represents the applicable step of the interaction procedure. As stated at the head of this section, Ramsey resonance is observed through three steps: irradiation of the electromagnetic wave for time  $\tau$ , suspension of the electromagnetic wave for time  $T$ , and irradiation of the electromagnetic wave again for time  $\tau$ . Thus,  $k$  can take a value of 1 to 3. Let us remember that we are now assuming that the approximation of Eq. (24) holds when there is an interaction with the electromagnetic wave. Then, for  $k = 1$  and  $k = 3$ , Eqs.(36) and (37) can be approximated as

$$i \frac{\partial}{\partial \theta} \begin{pmatrix} \gamma_1 \\ \gamma_2 \end{pmatrix} = \begin{pmatrix} 0 & \frac{b}{2} \\ \frac{b}{2} & 0 \end{pmatrix} \begin{pmatrix} \gamma_1 \\ \gamma_2 \end{pmatrix} \quad (40)$$

The solution of this equation is expressed in the matrix expression as follows:

$$M^{(1)}(\theta) = M^{(3)}(\theta) = \exp -i \begin{pmatrix} 0 & \frac{b\theta}{2} \\ \frac{b\theta}{2} & 0 \end{pmatrix} \quad (41)$$

When the even and odd-order terms of the expanded power series are separated and rearranged using

$$\begin{pmatrix} 0 & -i \\ -i & 0 \end{pmatrix}^2 = \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix} \quad (42)$$

the result is the following expression:

$$M^{(1)} = M^{(3)} = \begin{pmatrix} \cos \frac{b\tau}{2} & -i \sin \frac{b\tau}{2} \\ -i \sin \frac{b\tau}{2} & \cos \frac{b\tau}{2} \end{pmatrix} \quad (43)$$

For  $k = 2$ , there is no electromagnetic wave to interact with, thus  $b = 0$ , which changes Eqs.(36) and (37) into the following form:

$$i \frac{\partial}{\partial \theta} \begin{pmatrix} \gamma_1 \\ \gamma_2 \end{pmatrix} = \begin{pmatrix} \frac{\Omega_0}{2} & 0 \\ 0 & -\frac{\Omega_0}{2} \end{pmatrix} \begin{pmatrix} \gamma_1 \\ \gamma_2 \end{pmatrix} \quad (44)$$

The diagonal matrix can be integrated immediately and the solution is

$$M^{(2)} = \begin{pmatrix} \exp -\frac{i\Omega_0 T}{2} & 0 \\ 0 & \exp \frac{i\Omega_0 T}{2} \end{pmatrix} \quad (45)$$

Using the results obtained above, assuming an initial state as  $c_1 = 1$  and  $c_2 = 0$ , the state obtained after irradiating the electromagnetic wave for time  $\tau$ , waiting for time  $T$ , and irradiating the electromagnetic wave again for time  $\tau$  is calculated as

$$\begin{pmatrix} \gamma_1(\tau + T + \tau) \\ \gamma_2(\tau + T + \tau) \end{pmatrix} = M^{(3)}(\tau) M^{(2)}(T) M^{(1)}(\tau) \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad (46)$$

Substituting Eqs.(43) and (45) into this equation gives the following result:

$$\gamma_2(\tau + T + \tau) = -i \sin b\tau \cos \frac{\Omega_0 T}{2} \quad (47)$$

Using this, the probability is calculated with

$$P(\tau) = |\gamma_2(\tau + T + \tau)|^2 \quad (48)$$

to give

$$P(\tau) = \frac{1}{2} \sin^2 b\tau (1 + \cos \Omega_0 T) \quad (49)$$

which has already been presented.

As shown here, when there is a diagonal Hamiltonian corresponding to the energy levels and there is also a non-diagonal weak perturbation, then if the atoms are prepared to take the same state, the application of the perturbation twice (with a period of time between applications) generates a cosine term whose phase is proportional to the product of the

time interval and the difference between the perturbation frequency and the resonance frequency. This is Ramsey resonance. In this case the difference between the perturbation frequency and the resonance frequency can be measured with extremely high sensitivity by making the time interval for the perturbation large.

The result obtained in Eq. (49) is only valid under the approximation of Eq. (24). The result gradually changes as the difference between the perturbation frequency and the resonance frequency increases. Further, in practice, the atoms in the atomic beam have various velocities, and the transition probability is expressed as

$$P = \frac{1}{2} \int_0^{\infty} f(\tau) \sin^2 b \tau [1 + \cos(\omega - \omega_0)T] d\tau \quad (50)$$

Here,  $f(\tau)$  represents the normalized interaction time for the atom. For thermal beam standards, if the velocity distribution function  $p(v)$  is given, noting that  $v = l/\tau$ , this normalized interaction time is expressed as

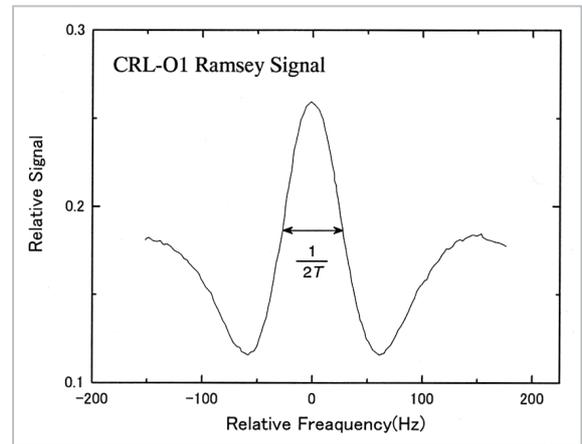
$$f(\tau) = \frac{l}{\tau^2} P\left(\frac{l}{\tau}\right) \quad (51)$$

In this way, the Ramsey resonance signal actually observed does not behave exactly as expressed in Eq. (49) for various reasons, except when  $\omega$  is extremely close to  $\omega_0$ . Figure 6 shows the Ramsey resonance signal obtained from our optically pumped primary frequency standard CRL-O1.

## 5 Conclusion

Atomic frequency standards are one of the most precise measurement instruments in use, measuring the quantities of time and frequency with significant figures of 14 to 15 orders of magnitude.

The principles of operation of these stan-



**Fig.6** Ramsey resonance signal obtained from the CRL-O1 optically pumped primary frequency standard

dards are based on extremely advanced modern physics, including electromagnetics and quantum mechanics. I recognize that the phenomena studied in modern physics have been tested strictly, and obtained greater reliability, in turn leading to the exceptional accuracy of atomic frequency standards. In this report I have attempted to describe some of the elementary physics required to understand the operational principles involved.

The topics discussed here are limited to atomic energy levels, emissions and lifetime in quantum transitions, and the Ramsey resonance, but even this limited discussion leaves out a great deal. I hope nevertheless that it will be of some help in permitting the reader to grasp the phenomena intuitively, to gain an understanding of approximate quantitative evaluation, and to pursue more detailed research on the subject. Please see Reference [14] for a description of the structure of the cesium primary frequency standards and of the various frequency shift factors. I hope later to have the opportunity to address these and other issues that could not be discussed here, as well as to introduce a more advanced and detailed description of the relevant phenomena.

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### 【Apology and correction】

In this paper, a misprint of the unit in a figure was found in "2 Atomic spectra and energy levels." The misprint appeared in a lower part of Fig.2 (Energy levels of the cesium atom: fine and hyperfine structures), which is on the right-hand side in Page 35. The erratum in the figure is already corrected.

(error) 9 192 631 770MHz → (correct) 9 192 631 770Hz

We sincerely apologize for that erratum and take this opportunity for correction. (October 31, 2017)