

## Rotational Frequency Shift

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(Received 5 November 1996)

The notion of the rotational frequency shift, an analog of the Doppler shift, is introduced. This new frequency shift occurs for atomic systems that lack rotational invariance, but have stationary states in a rotating frame. The rotational frequency shift is given by the scalar product of the angular velocity and the angular momentum of the emitted photon in full analogy with the standard Doppler shift which is given by the scalar product of the linear velocity of the source and the linear momentum of the photon. The rotational frequency shift can be observed only in a Mössbauer-like regime when the angular recoil is negligible. [S0031-9007(97)02782-8]

PACS numbers: 32.70.Jz, 06.30.Ft, 32.90.+a, 41.60.-m

The purpose of this Letter is to describe a new effect: the frequency shift of emitted (or absorbed) photons that is due to the rotation of the radiating system. This rotational frequency shift (RFS) is a close analog of the standard, first-order Doppler shift (the latter might be called a translational frequency shift) but there are two important differences.

First, the dynamical laws are invariant under a uniform translation, but they are not invariant under a uniform rotation. Therefore, the differences between the energy levels are not changed by a uniform translation but they are dynamically modified by a uniform rotation, owing to the centrifugal and Coriolis forces. Hence, the frequency of a photon emitted (absorbed) by a system moving with constant velocity is modified *only* by the Doppler shift, while in the case of a rotating system the photon frequency is modified *both* by the changes in the energy levels and by the RFS. For rotationally invariant systems, these two effects completely cancel each other as a result of the conservation of angular momentum, and the observed photon frequency is unchanged, as one would have anticipated.

Second, under normal conditions the recoil corrections are much more significant for the RFS than for the Doppler shift. In order to observe the RFS one must work in a Mössbauer-like regime. The atomic system must be embedded in a larger structure that will provide the angular momentum of the emitted photon.

The RFS should not be confused with the ordinary linear Doppler shift observed for rotating objects (for example, stars or galaxies) that is due to the instantaneous linear motion of the emitter. This linear Doppler shift is maximal in the plane of rotation while the RFS is maximal along the angular velocity, that is in the direction perpendicular to the instantaneous velocity. Thus the RFS competes with the quadratic Doppler shift rather than with the linear Doppler shift.

Our discussion will be based on the nonrelativistic Schrödinger equation for the atomic system with a time

dependent Hamiltonian. The photon emission will be treated in the first order of perturbation theory. Our derivation of the RFS will be done in parallel with the calculation of the standard Doppler shift in order to exhibit their similarities and differences.

As a generic model of a radiating system we shall consider an electron bound by a potential  $V(t)$  and interacting via minimal coupling with the quantized electromagnetic field. We assume that the time dependence of the potential is of the form  $V(t) = V(\mathbf{r}(t))$ . For a uniform translation and a uniform rotation,  $\mathbf{r}(t)$  is given by

$$\text{Translation } x(t) = x, \quad y(t) = y, \quad z(t) = z - vt, \quad (1)$$

$$\begin{aligned} \text{Rotation } x(t) &= x \cos(\Omega t) + y \sin(\Omega t), \\ y(t) &= -x \sin(\Omega t) + y \cos(\Omega t), \\ z(t) &= z. \end{aligned} \quad (2)$$

Since the time dependence of the potential is prescribed, our analysis will be applicable only to those cases when the change in the uniform motion of the potential caused by the photon emission (absorption) can be disregarded. This means that we neglect all recoil corrections.

The time dependence of the state vector  $\Psi(t)$  is determined by the evolution equation (in atomic units)

$$\begin{aligned} i \partial_t \Psi(t) &= H(t) \Psi(t) \\ &= \left[ \frac{1}{2} \mathbf{p}^2 + V(t) + H_F + H_I \right] \Psi(t), \end{aligned} \quad (3)$$

where  $H_F$ ,

$$\begin{aligned} H_F &= \frac{1}{2} \int d^3x (\mathbf{D}^2/\epsilon_0 + \mathbf{B}^2/\mu_0) \\ &= \sum_{\lambda} \int d^3k \omega(\mathbf{k}) a_{\lambda}^{\dagger}(\mathbf{k}) a_{\lambda}(\mathbf{k}) \\ &= \sum_{JM\lambda} \int d\omega \omega a_{JM\lambda}^{\dagger}(\omega) a_{JM\lambda}(\omega), \end{aligned} \quad (4)$$

is the free field Hamiltonian and  $H_I$ ,

$$H_I = \mathbf{p} \cdot \mathbf{A}(\mathbf{r}) + \frac{1}{2} \mathbf{A}^2(\mathbf{r}), \quad (5)$$

is the interaction Hamiltonian.

In order to determine the spectrum of emitted photons, we construct the transition amplitude  $A_{fi}$ ,

$$A_{fi} = \langle \Psi_f | U(t, t_0) | \Psi_i \rangle. \quad (6)$$

The evolution operator  $U(t, t_0)$  satisfies the equation

$$i \partial_t U(t, t_0) = H(t) U(t, t_0), \quad (7)$$

and the initial condition  $U(t_0, t_0) = 1$ . Next, we introduce a unitary transformation  $\exp(iGt)$  to get rid of the time dependence of the Hamiltonian  $H(t)$ , transforming it back to time  $t = 0$ ,

$$H(0) = e^{iGt} H(t) e^{-iGt}. \quad (8)$$

The operators  $G$  are the generators of uniform translations and uniform rotations,

$$\text{Translation } G = v P_z, \quad (9)$$

$$\text{Rotation } G = \Omega M_z, \quad (10)$$

where  $P_z$  and  $M_z$  denote the operators of the  $z$  component of the *total* linear momentum and angular momentum (including the electromagnetic field),

$$P_z = p_z + \sum_{\lambda} \int d^3k k_z a_{\lambda}^{\dagger}(\mathbf{k}) a_{\lambda}(\mathbf{k}), \quad (11)$$

$$M_z = x p_y - y p_x + \sum_{JM\lambda} \int d\omega M a_{JM\lambda}^{\dagger}(\omega) a_{JM\lambda}(\omega). \quad (12)$$

Note that the Hamiltonians  $H_F$  and  $H_I$  are invariant under translations and rotations and, therefore, they are not changed by the unitary transformations (8). One may check that the solution of the evolution equation (7) satisfying the initial condition has the form

$$U(t, t_0) = e^{-iGt} e^{-i(H_0+H_I)(t-t_0)} e^{iGt_0}, \quad (13)$$

where

$$H_0 = \frac{1}{2} \mathbf{p}^2 + V(0) + H_F - G. \quad (14)$$

In the first order of perturbation theory, we obtain

$$U(t, t_0) \approx e^{-iGt} e^{-iH_0(t-t_0)} e^{iGt_0} - i e^{-iGt} \int_{t_0}^t d\tau e^{-iH_0(t-\tau)} \times \mathbf{p} \cdot \mathbf{A}(\mathbf{r}) e^{iH_0(t_0-\tau)} e^{iGt_0}. \quad (15)$$

Upon inserting this expression into the formula (6) for the transition amplitude, we get

$$A_{fi} \approx \langle \Phi_f | e^{-iH_0(t-t_0)} | \Phi_i \rangle - i \langle \Phi_f | \int_{t_0}^t d\tau e^{-iH_0(t-\tau)} H_I e^{iH_0(t_0-\tau)} | \Phi_i \rangle, \quad (16)$$

where

$$|\Phi_i\rangle = e^{iGt_0} |\Psi_i\rangle, \quad |\Phi_f\rangle = e^{iGt} |\Psi_f\rangle. \quad (17)$$

The transition amplitude (16) will lead to the Fermi Golden Rule for the transition rate provided the auxiliary vectors  $|\Phi_{i,f}\rangle$  are chosen as eigenvectors of  $H_0$ . In order to study spontaneous emission, we shall assume that the initial state comprises the excited state of the electron and the vacuum state of the field and the final state comprises the ground state of the electron and a one photon state. The corresponding eigenvectors  $|\Phi_{i,f}\rangle$  have a product form and they satisfy the following eigenvalue equations:

$$H_0 |\phi_e(\mathbf{r})\rangle |0\rangle = E_e |\phi_e(\mathbf{r})\rangle |0\rangle, \quad (18)$$

$$H_0 |\phi_g(\mathbf{r})\rangle |1_{ph}\rangle = (E_g + \omega - \Delta) |\phi_g(\mathbf{r})\rangle |1_{ph}\rangle, \quad (19)$$

where  $\phi_e(\mathbf{r})$  and  $\phi_g(\mathbf{r})$  are the eigenfunctions of the electronic part  $H_E$  of  $H_0$ ,

$$\text{Translation } H_E = \frac{1}{2} \mathbf{p}^2 + V(0) - v p_z, \quad (20)$$

$$\text{Rotation } H_E = \frac{1}{2} \mathbf{p}^2 + V(0) - \Omega(x p_y - y p_x), \quad (21)$$

corresponding to the eigenvalues  $E_e$  and  $E_g$ . The shift  $\Delta$  is an eigenvalue of the photonic part of the generator  $G$  [cf. Eqs. (11) and (12)] and is equal to  $v k_z$  in the case of translation or to  $\Omega M$  in the case of rotation. The vectors  $|\Phi_{i,f}\rangle$  represent stationary states; the electronic probability densities  $|\phi_{e,g}(\mathbf{r})|^2$  are time independent. The original vectors  $|\Psi_{i,f}\rangle$ , related to the eigenvectors  $|\Phi_{i,f}\rangle$  by the transformation  $\exp(-iGt)$  represent nonstationary states. Since the transformation  $\exp(-iGt)$  acting on the electronic wave function replaces its argument  $\mathbf{r}$  by  $\mathbf{r}(t)$ , the electronic wave functions depend on time not only through a phase factor  $\exp(-iEt)$  but also through  $\mathbf{r}(t)$ . These states may be called quasistationary since the corresponding electronic probability densities  $|\psi_{e,g}(\mathbf{r}(t))|^2$  undergo a uniform motion according to the formulas (1) and (2).

The spectrum of the emitted photons can be determined from the standard formula for the decay rate  $w_{if}$  applied to the amplitude (16)

$$w_{if} = 2\pi |\langle \Phi_f | H_I | \Phi_i \rangle|^2 \delta(E_e - E_g - \omega + \Delta). \quad (22)$$

Hence the frequency of the emitted photon is shifted by  $\Delta$ . For a uniform translation,  $\Delta$  represents the standard Doppler shift and for a uniform rotation,  $\Delta$  represents the *rotational frequency shift*,

$$\text{Translation (Doppler)} \quad \omega = E_e - E_g + v k_z, \quad (23)$$

$$\text{Rotation (RFS)} \quad \omega = E_e - E_g + \Omega M. \quad (24)$$

In the case of a uniform translation the electronic energy difference  $E_e - E_g$  is exactly the same as for a system at rest because the electronic part of the Hamiltonian  $H_0$  is unitarily equivalent (up to a constant) to the corresponding Hamiltonian at rest

$$e^{ivz} \left[ \frac{1}{2} \mathbf{p}^2 + V(0) - v p_z \right] e^{-ivz} = \frac{1}{2} \mathbf{p}^2 + V(0) - \frac{1}{2} v^2. \quad (25)$$

Therefore for a uniform translation the only change in the spectrum of the emitted photons is the Doppler shift. This is simply the result of the Galilean invariance of nonrelativistic quantum mechanics. The situation is quite different for a uniform rotation. The spectrum of the photons emitted by a rotating source is not only shifted by  $\Delta = \Omega M$  but it is also, in general, modified due to changes of the energy differences  $E_e - E_g$ .

This modification is especially simple for rotationally invariant potentials when  $M_z$  commutes with the Hamiltonian  $H_0$ . In this case the wave functions  $\phi_e(\mathbf{r})$  and  $\phi_g(\mathbf{r})$  can be chosen as eigenfunctions of the  $z$  component of the electronic angular momentum belonging to certain eigenvalues  $m_e$  and  $m_g$ . Then the change in the energy difference caused by the rotation is equal to  $\Omega(m_e - m_g)$ . This change cancels *completely* the RFS owing to the conservation of angular momentum and no shift is observed in the laboratory frame. Therefore, the RFS cannot be seen in isolated atoms but it might occur, in principle, in isolated molecules or in atoms placed in an environment that destroys the rotational symmetry.

Even when the potential is not rotationally invariant, the consequences of the RFS are not always significant. In particular, when the rotating body is of macroscopic dimensions the RFS is much smaller, in general, than other effects. For example, in the experiment [1] designed to measure the transverse Doppler shift the emitter and the absorber were mounted on a rotating disk. Even though the angular velocity was large enough to observe the quadratic Doppler shift, the RFS calculated for the conditions of this experiment could not have been seen (it is only about  $10^{-4}$  of the quadratic Doppler shift). The RFS is so small in this case because the ratio of the wavelength to the linear dimensions of the rotating system is exceedingly small. The RFS might be seen for macroscopic rotating systems when the emitted radiation is in the radio-wave range, as is the case for transitions involving nuclear magnetic moments. Taking the frequency of such a transition in the range of  $10^8$  Hz and assuming an ultrafast centrifuge rotating at  $10^3$  Hz, one obtains the RFS shift of the order of  $10^{-5}$  of the resonance frequency. Such shifts might be detectable. In order to observe the RFS in the optical domain, the rotational frequency must be much higher and that can be achieved only when the rotating systems are of atomic dimensions.

The simplest model of a rotating atomic system with broken rotational symmetry is an electron bound by a rotating asymmetric harmonic potential in two dimensions,

$$V(t) = \frac{1}{2} [\mu x^2(t) + \nu y^2(t)], \quad (26)$$

where  $\mu$  and  $\nu$  are two arbitrary real parameters and the dependence of  $x$  and  $y$  on time is given by the formulas (2). An especially interesting case is when one of the two oscillators is inverted (one of the parameters  $\mu$  or  $\nu$  is negative). Such a model has been recently used to predict the existence and to describe the main features of Trojan wave packets of Rydberg electrons in atoms [2] and in

rotating molecules with large electric dipole moments [3]. Since all more elaborate numerical calculations [4–7] fully confirmed the validity of the estimates based on the harmonic approximation, we shall use the model with the potential (26) to study the significance of the RFS in realistic situations.

The Trojan states of Rydberg electrons in hydrogenic atoms are described by nonspreading localized wave packets circling on a large orbit around the nucleus. Their stability is due to an interplay between the centrifugal force and the Coriolis force acting in the rotating frame and the electric field of a circularly polarized electromagnetic wave propagating in the direction perpendicular to the orbit. They were named by us the Trojan states because their mathematical description is similar to the description of the orbits of Trojan asteroids in the Sun-Jupiter system [2]. The Trojan states of electrons offer a perfect example of an atomic system that will exhibit the RFS since in the laboratory frame the interaction with the electric field of the wave is described (in the dipole approximation) by the rotating potential  $V(t) = \mathcal{E}[x \cos(\Omega t) + y \sin(\Omega t)]$ . The description of the Trojan states in the harmonic approximation [2] leads to the Hamiltonian in which the parameters  $\mu$  and  $\nu$  are determined by the frequency of the wave  $\Omega$  and by the parameter  $q$  that measures the ratio between the Coulomb force and the centrifugal force,  $\mu = -2q\Omega^2$ ,  $\nu = q\Omega^2$ . The frequencies  $\omega_{\pm}$  in this case are

$$\omega_{\pm} = \Omega \sqrt{(2 - q \pm \sqrt{9q^2 - 8q})/2}. \quad (27)$$

They are real for  $8/9 \leq q \leq 1$ .

We shall use now the harmonic oscillator model to exhibit the main features of the RFS for realistic atomic systems. The electronic part  $H_E$  of the time independent Hamiltonian for the potential (26) is in this case

$$H_E = \frac{1}{2} \mathbf{p}^2 + \frac{1}{2} (\mu x^2 + \nu y^2) - \Omega (x p_y - y p_x). \quad (28)$$

This Hamiltonian describes two harmonic oscillators with the following characteristic frequencies

$$\omega_{\pm} = \sqrt{(\mu + \nu + 2\Omega^2 \pm \sqrt{(\mu - \nu)^2 + 8(\mu + \nu)\Omega^2})/2} \quad (29)$$

that determine the spacing between the energy levels. These frequencies are real when the parameters  $\mu$ ,  $\nu$ , and  $\Omega$  satisfy the conditions

$$\mu \geq \Omega^2 \quad \text{and} \quad \nu \geq \Omega^2, \quad (30)$$

or

$$\begin{aligned} -3\Omega^2 \leq \mu \leq \Omega^2, \quad -3\Omega^2 \leq \nu \leq \Omega^2, \\ \text{and} \quad (\mu - \nu)^2 + 8(\mu + \nu)\Omega^2 \geq 0. \end{aligned} \quad (31)$$

In a degenerate case of an isotropic potential, when  $\mu = \nu$ , the frequencies  $\omega_{\pm}$  are linear functions of the angular velocity  $\omega_{\pm} = \sqrt{\mu} \pm \Omega$ . This is a special example of a rotationally invariant potential, when the RFS is exactly

canceled by the dynamical modification of the electronic energy difference. For anisotropic potentials the energy differences are nonlinear functions of  $\Omega$  and a complete cancellation between the RFS and the dynamical modifications of the energy differences is impossible.

The RFS that occurs in the emission of photons during transitions between the atomic Trojan states has dramatic consequences. The frequencies (24) of the photons emitted during the transitions between the neighboring energy levels of the oscillators, as seen by the observer in the laboratory frame, are

$$\omega = \omega_{\pm} + \Omega M. \quad (32)$$

The observed photon frequency is drastically shifted by the RFS; the shift *exceeds* the original transition frequency in the rotating frame. This huge effect is due to the fact that the rotation is not just a small modification of the dynamics, but the mere existence of the Trojan states depends on it. Since both frequencies  $\omega_{\pm}$  are smaller than  $\Omega$ , the RFS has a very large effect in this case. The emission of photons with  $M = -1$  is forbidden. This means that the photons emitted spontaneously by the Trojan electrons have the same projection of the angular momentum on the  $z$  axis as the photons constituting the strong wave that is driving the system. At this point we would like to stress that the RFS and the transfer of the angular momentum to the emitted photon must not in any direct way be attributed to the photons that make the circularly polarized wave. The same frequency shift is obtained for all rotating potentials, regardless of their nature. For example, it would appear for nonspherical rotating nuclei, provided one can treat the rotating nuclear potential as fully prescribed.

In order to illustrate these general results with specific numbers, we shall consider a hydrogen atom driven by the circularly polarized wave with the frequency of 200 GHz and the field amplitude of 1930 V/m, the same as in Ref. [2]. In this case, the separations between the neighboring oscillator levels in the rotating frame are  $1.92 \times 10^{11}$  Hz and  $0.66 \times 10^{11}$  Hz. The frequencies of the circularly polarized radiation observed in the laboratory frame will be shifted by  $2 \times 10^{11}$  Hz and they will be equal to  $3.92 \times 10^{11}$  Hz and  $2.66 \times 10^{11}$  Hz, respectively. Both frequencies are quite different from the frequency of the driving wave.

Our derivation of the RFS depends crucially on the validity of the no-recoil approximation. This assumption is satisfied very well for the Doppler shift but the effect of recoil in the rotational motion is much more significant than in the translational motion. Usually, one may disregard the perturbation of the translational motion by the emission of the photon because the relative change in the velocity is of the order of  $10^{-4}$ . In contrast, the relative change of the angular velocity of a molecule caused by an emission of a photon, as argued below, is several orders of magnitude

larger. The principle of energy equipartition gives the relation between the angular velocity and the linear velocity  $\Omega \approx v/R$ , where  $R$  is the effective radius of the molecule. Therefore the relative changes of the angular velocity and the linear velocity are related by a large factor, the ratio of the wave length to the molecular radius,

$$\delta\Omega/\Omega \approx (1/kR)(\delta v/v), \quad (33)$$

where  $k$  is the photon wave vector. This means that for an isolated molecule the angular recoil should be taken into account. The whole molecule, not just the electron undergoing the radiative transition, must be considered as a radiating system. For the molecule, treated as a closed system, the RFS is unobservable because the total angular momentum is conserved and we end up in the category of rotationally invariant systems. However, for strongly driven atoms or molecules, as in the case of atomic Trojan states, a Mössbauer-like regime is reached since the external field sustains the uniform rotational motion. Under such conditions, the angular recoil can be disregarded since the angular momentum of the emitted photon is provided by the macroscopic field.

The standard explanation of the Doppler shift is based on the kinematics of a special theory of relativity [8,9]. The RFS may also be explained in a similar vein but one must apply general theory of relativity because the rotating coordinate frame is not inertial. One may use in this case the known transformation properties of the Maxwell equations under general coordinate transformations to relate the frequency of the photon emitted in the rotating frame to the frequency observed in the laboratory frame. However, the RFS is always accompanied by the modifications of the emitter dynamics due to rotation and the net frequency shift can be observed *only for quantum mechanical systems* when the electronic Hamiltonian does not commute with the generator of rotations.

This work has been supported by the KBN Grant No. 2P30B01309.

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