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Selection rules and centre-of-mass motion of ultracold atoms

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Abstract. In recent years much attention has been paid to the quantized evolution of the centre-of-mass momentum and position of ultracold atoms in light fields. We consider the effects resulting from the quantization of the external angular momentum variables. We investigate how spin and orbital angular momentum of light are transferred to internal and external angular momentum of an atom in dipole and quadrupole transitions.

1. Introduction

When a light field interacts with an atom, one can usually neglect the centre-of-mass motion of the atom, and consider the effects on the internal electronic motion only. It suffices, e.g., to specify the change in energy and angular momentum of the electron in the centre-of-mass system.

For a cold atom, however, the recoil effect of even a single photon can no longer be neglected: a single photon can change the external state of the atom. Even in this case the external atomic motion can in general still be described classically. Only when the atom is cooled down further to the recoil temperature, and its de Broglie wavelength λ_B becomes comparable to the wavelength λ of the light, must the external motion be quantized [1].

The prime example of a pure quantum effect arising from the quantized external motion of an ultracold atom is the effect of velocity-selective coherent population trapping [2]. An atom in a light field gets trapped in a state that is insensitive to the light, and which is a superposition of two states with different external momenta, differing by two photon momenta. A cooling scheme based on this mechanism has been shown to lead to temperatures below the recoil temperature [2]. This idea has been extended to three dimensions [3] and to different atomic transitions [4].

Another example is the quantized motion of cold atoms in optical potential wells with the size of a wavelength, produced by two counterpropagating laser beams [5]. Here atoms can be trapped in a single well and can occupy a single quantized energy level therein. Also the occurrence of tunnelling from one well to an adjacent one has been predicted [6]. The presence of the discrete energy levels and the localization of an atom in one well have been observed experimentally [7, 8]. Recently cooling of atoms and their quantized motion in optical wells has also been observed in two- and three-dimensional laser beam configurations [9–11].

Now photons also carry angular momentum [12]. Therefore, photons can exert, in addition to a force, also a torque on an atom. There is one important difference, however:

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the momentum of the photon is always transferred to the *external* momentum of the atom, whereas the photon angular momentum in general is transferred to the *internal* atomic angular momentum. In fact, the well known dipole selection rules $\delta l = \pm 1$ refer exclusively to the internal electronic state, and are understood as arising from the spin angular momentum of the photon, which is equal to 1. Recently it has been shown that also orbital angular momentum of light, arising from the transverse spatial dependence of a light beam, is a meaningful concept, accessible to experimental verification [13–16]. This quantity will in general be transferred to the *external* angular momentum of an atom [17]. This implies that it is the *orbital* angular momentum of light that can make atoms rotate around a given point or axis.

Some questions remain: what effects arise for ultracold atoms, when the external angular momentum must be quantized? What happens in a quadrupole transition, where $\delta l = \pm 2$? Does the second unit of angular momentum come from the orbital angular momentum of the photon? In order to answer these questions, we investigate the selection rules concerning (internal and external) angular momenta during the absorption or emission of a photon by an atom. We wish to clarify the conservation laws underlying these rules, and in particular the role of the centre-of-mass motion of the atom. We will discuss some unfamiliar effects on cold atoms resulting from angular momentum transfer between photons and atoms.

Throughout this paper we will use the long-wavelength approximation, i.e. we assume that the wavelength λ of the light is large compared to the size a of the atomic system under consideration. This justifies making an expansion in the small parameter a/λ. It should be noted that this approximation does not imply any restriction on the de Broglie wavelength of the atom: λ_B and a are independent atomic quantities, determined by the size of the external and of the internal part of the wavefunction, respectively. This paper is organized as follows. In order to be able to discuss conservation laws of angular momentum one needs to define electromagnetic field modes with well defined angular momenta. Several possible definitions are reviewed in section 2. In section 3 we examine in some detail the selection rules for photon absorptions and emissions by an atom. Emphasis is put on the modifications that arise due to the inclusion of the centre-of-mass motion of the atom. Some explicit examples are presented in section 4. They serve to show how spin and orbital angular momenta of a photon are distributed over the internal and external angular momenta of the atom, and why the temperature plays an important role. The results are summarized and discussed in section 5. Finally, in the appendix we define the internal and external variables as used in this paper.

2. Photons and angular momentum

We first establish the notation and definitions for the well known multipole waves [12, 18], in which photons are in eigenstates of the operators for total field angular momentum J^2 and its projection J_z. In the second subsection, we define photons in eigenstates of the projected 'orbital' and 'spin' angular momenta L_z and S_z [17]. Finally, we will briefly discuss Laguerre–Gaussian modes, which have been shown to be producable by a transformation of laser beams [15], and which also possess well defined S_z and L_z.

A photon is here defined as an elementary excitation of a field mode. A field mode is represented by a mode function $\boldsymbol{F}_\lambda(\boldsymbol{r})$, which is a transverse vector solution to the wave equation with wave number $k_\lambda = \omega_\lambda/c$. Each mode function can be chosen as the eigenfunction of a prescribed set of four commuting Hermitian operators. The mode is then

Table 1. Notation for the quantum numbers related to the various angular momenta of photons and atoms.

Species	Operator	Quantum number	Eigenvalue
Photon	J^2	j	$\hbar^2 j(j+1)$
Photon	J_z	m	$m\hbar$
Photon	S_z	s	$s\hbar k_z/k$
Atom	J^2_{int}	l	$\hbar^2 l(l+1)$
Atom	$J_{\text{int},z}$	m	$m\hbar$
Atom	J^2_{ext}	L	$\hbar^2 L(L+1)$
Atom	$J_{\text{ext},z}$	M	$M\hbar$
Atom	J^2_{total}	J	$\hbar^2 J(J+1)$

specified by the four eigenvalues of these operators, denoted collectively by λ, which are at the same time the quantum numbers of the photons from that field mode.

Once we have thus defined a complete set of normalized transverse vector functions $\boldsymbol{F}_\lambda$, the operator $\boldsymbol{A}$ for the vector potential in the Coulomb gauge can be expanded in this set as [19]

$$\boldsymbol{A}(\boldsymbol{r}) = \sum_\lambda \sqrt{\frac{\hbar}{2\epsilon_0 \omega_\lambda}} [a_\lambda \boldsymbol{F}_\lambda(\boldsymbol{r}) + a_\lambda^\dagger \boldsymbol{F}_\lambda^*(\boldsymbol{r})] \tag{1}$$

where a_λ and $a_\lambda^\dagger$ are the annihilation and creation operators for photons in the mode λ. In the examples we will consider, two of the four quantum numbers will refer to angular momentum variables. The notation for these variables is summarized in table 1.

2.1. Multipole waves

We review here some well known results concerning the multipole waves, as described in [12, 19]. One constructs eigenstates of the four commuting Hermitian operators for total angular momentum J^2, its projection along the z axis J_z, energy, and parity. The set of quantum numbers is correspondingly given by $\lambda = (j, m, \omega, P)$, where the parity P takes the values ± 1, and where ω is the frequency. There are two types of waves: electric multipole waves with parity $P = (-1)^j$ and magnetic multipole waves with opposite parity $P = (-1)^{j+1}$. The explicit expressions for the mode functions can be found in [12], or, for the corresponding electric and magnetic fields in [19]. The only multipole fields that are non-vanishing in the origin are the electric dipole waves with $j = 1$ and $m = \pm 1, 0$.

2.2. Bessel waves

One can also construct field modes as the eigenfunctions of the commuting set of operators for the projection of the angular momentum J_z, linear momentum P_z, energy and 'spin' S_z. The corresponding quantum numbers are $\lambda = m, k_z, \omega, s$, where $\hbar k_z$ is the momentum in the z direction, and where $s = \pm 1$ denotes the polarization or helicity (right or left hand). Only for waves propagating in the z direction, i.e. for waves with $k_z = \pm k$, is the quantum operator S_z a true spin angular momentum operator [17, 20]. In that case it has discrete eigenvalues $\pm\hbar$. Explicit expressions for the corresponding mode functions $\boldsymbol{F}_\lambda$ were derived in [17]. The result takes the form (the $\pm$ refers to the polarization index s)

$$\frac{1}{\sqrt{2}}(F_x + \mathrm{i}F_y) = \frac{k_z \mp k}{2k} f(k_t, k_z, m+1)$$

$$\frac{1}{\sqrt{2}}(F_x - iF_y) = \frac{k_z \pm k}{2k} f(k_t, k_z, m-1)$$
$$F_z = \frac{k_t}{\sqrt{2}k} f(k_t, k_z, m) \tag{2}$$

where the functions $f(k_t, k_z, m)$ are defined in cylindrical coordinates (ρ, z, ϕ) as

$$f(k_t, k_z, m) = J_m(k_t\rho)\exp(ik_z z)\exp(im\phi)/N. \tag{3}$$

Here J_m is the mth-order Bessel function, $\hbar k_t$ is the momentum in the direction perpendicular to the z direction: $k_t^2 = k^2 - k_z^2$, and N is a normalization constant. Note that these modes do not have well defined total angular momentum J^2, nor a definite parity. The only fields that are non-vanishing in the origin are those with $m = \pm 1, 0$.

These waves generalize so-called Bessel beams, which are proportional to the zeroth-order Bessel functions, and which have been produced recently to moderately high intensity using a specific zone plate configuration [21].

2.3. *Laguerre–Gaussian beams*

Laguerre–Gaussian (LG) beams are special solutions to the paraxial equation [22]. They belong, therefore, to a class of exact solutions to an approximate equation describing light beams with a well defined propagation direction. The LG modes are cylindrically symmetric beams, with an azimuthal dependence given by $\exp(im\phi)$. They can be viewed as superpositions of Bessel waves with fixed value of m and with different $k_t \ll k$. These beams can be produced from (laser) Hermite–Gaussian beams by using a special configuration of astigmatic cylindrical lenses [13–15]. Thus one has been able to produce modes with up to three units of orbital angular momentum L_z per photon [15].

3. Selection rules

We review here how the selection rules for photon absorption and emission processes arise in the case that the atomic centre of mass motion is neglected. For more details see [18]. Subsequently we include this external motion, and investigate the resulting modifications of the selection rules and conservation laws.

3.1. *Centre of mass is neglected*

The atom is modelled by a spinless electron with coordinate $\boldsymbol{r}$ and momentum $\boldsymbol{p}$, charge $-e$ and mass μ that is bound to the origin by a given potential $V(r)$. The electron is confined by this potential to move in a region $|\boldsymbol{r}| < a$. We neglect the centre-of-mass motion, effectively assuming the atom to have an infinitely heavy nucleus. The centre-of-mass position $\boldsymbol{R}$ is not a dynamical variable, but its value is fixed and equal to $\boldsymbol{R} = 0$.

The electron interacts with an external electromagnetic field, which is represented by the vector potential $\boldsymbol{A}$ in the Coulomb gauge. The interaction part of the Hamiltonian is, neglecting the $\boldsymbol{A}^2$ term†, given by

$$H_I = \frac{e}{\mu}\boldsymbol{A}(\boldsymbol{r})\cdot\boldsymbol{p}. \tag{4}$$

The wavelength of the light is assumed to be long, so that it is legitimate to expand in a/λ. This boils down to expanding the operator $\boldsymbol{A}(\boldsymbol{r})$ (i.e. the field modes F_λ) around the origin,

$$\boldsymbol{A}(\boldsymbol{r}) = \boldsymbol{A}(0) + (\boldsymbol{r}\cdot\nabla)\boldsymbol{A}(0) + \ldots. \tag{5}$$

† At the low intensities used in laser cooling this is certainly a valid approximation. Furthermore, in lowest order in a/λ, this term contributes only a (small) energy shift of all states.

3.1.1. Dipole transitions. The first term of the expansion yields the dipole interaction

$$H_I^{\mathrm{ED}} = \frac{e}{\mu}\boldsymbol{p}\cdot\boldsymbol{A}(0). \tag{6}$$

If the matrix element

$$\langle\psi_{\mathrm{f}}|H_I^{\mathrm{ED}}|\psi_{\mathrm{i}}\rangle$$

of this operator between some initial state $|\psi_{\mathrm{i}}\rangle$ and some final state $|\psi_{\mathrm{f}}\rangle$ is non-zero then the corresponding transition is an electric dipole transition. Let the initial atomic state be an eigenstate of total angular momentum J_{int}^2 and of the projection $J_{\mathrm{int},z}$ with eigenvalues $l(l+1)$ and m: $|\psi_{\mathrm{i}}\rangle = |l, m\rangle$. Then a dipole transition to a final state of the form $|\psi_{\mathrm{f}}\rangle = |l', m'\rangle$ is possible if and only if at least one the matrix elements

$$\langle l', m'|\boldsymbol{p}|l, m\rangle$$

is non-zero. This leads to the well known dipole selection rules [18] for $\delta m \equiv m' - m$ and for $\delta l \equiv l' - l$,

$$\delta m = \pm 1, 0 \qquad \delta l = \pm 1. \tag{7}$$

The usual interpretation of these rules is that they express angular momentum conservation, since the photon has spin 1. Indeed, if one makes an expansion of the field in multipole waves, then only one term has a non-vanishing value at the origin, namely the electric dipole field with total angular momentum $j = 1$, and projections $m = \pm 1, 0$. Thus, angular momentum is conserved, as only the electric dipole wave contributes to the dipole interaction (6). Parity is conserved as well, since the parity of an elctric dipole photon is odd, while the parity of the electron state changes sign.

3.1.2. Higher order transitions. The second term in the expansion (5), of first order in a/λ, yields two different types of interaction terms: first the magnetic dipole Hamiltonian of the form

$$H_I^{\mathrm{MD}} = \frac{e}{\mu}\overleftrightarrow{Q}_{\mathrm{MD}} : (\boldsymbol{r}\boldsymbol{p} - \boldsymbol{p}\boldsymbol{r}) \tag{8}$$

and second the electric quadrupole interaction

$$H_I^{\mathrm{EQ}} = \frac{e}{\mu}\overleftrightarrow{Q}_{\mathrm{EQ}} : (\boldsymbol{r}\boldsymbol{p} + \boldsymbol{p}\boldsymbol{r}). \tag{9}$$

We defined here

$$\overleftrightarrow{Q}_{\mathrm{MD}} = \tfrac{1}{2}([\nabla\boldsymbol{A}(0)]^t - \nabla\boldsymbol{A}(0)) \qquad \overleftrightarrow{Q}_{\mathrm{EQ}} = \tfrac{1}{2}([\nabla\boldsymbol{A}(0)]^t + \nabla\boldsymbol{A}(0)) \tag{10}$$

where the superscript t indicates the transpose of a tensor: $T_{ij}^t = T_{ji}$. The quantity $\overleftrightarrow{Q}_{\mathrm{MD}}$ is proportional to the magnetic field, $\overleftrightarrow{Q}_{\mathrm{EQ}}$ to the gradient of the electric field, both evaluated at the origin. A magnetic dipole transition is allowed if one of the matrix elements

$$\langle\psi_{\mathrm{f}}|(\boldsymbol{r}\boldsymbol{p} - \boldsymbol{p}\boldsymbol{r})|\psi_{\mathrm{i}}\rangle$$

is non-zero. The off-diagonal components of this tensor operator contain the cartesian components of the angular momentum operator, the diagonal part is a pure number. Neither operator can change the value of l, while only L_x and L_y can change m by one unit. Thus the selection rules are found to be [18]

$$\delta m = \pm 1, 0 \qquad \delta l = 0. \tag{11}$$

Analogously, one finds the electric quadrupole selection rules [18]

$$\delta m = \pm 2, \pm 1, 0 \qquad \delta l = \pm 2, 0. \tag{12}$$

In the multipole expansion the gradient of the field is non-vanishing at the origin for two kinds of waves: the magnetic dipole wave, with angular momentum 1, and the electric quadrupole wave with angular momentum 2, both with even parity. Hence, the selection rules (11) and (12) express, again, conservation of both angular momentum and parity. The usual comment is, that since the spin of the photon is 1, the second unit of angular momentum in a quadrupole transition must come from orbital angular momentum of the light field. In the next subsection we will show that this is not strictly true. Summarizing we note that in the multipolar expansion of the interaction

(i) in each different order of the expansion the atom interacts with a different multipole wave,

(ii) in lowest (electric dipole) order the spatial field dependence is neglected. Each higher order takes higher-order gradients of the field into account,

(iii) each expansion term of the interaction Hamiltonian is invariant under rotations around the origin. Therefore the selection rules express conservation of angular momentum of the corresponding multipole field and the electron,

(iv) external atomic angular momentum is excluded. The internal angular momentum change is determined completely by the angular momentum of the multipole wave.

3.2. Centre of mass is included

In this subsection we consider a bound system consisting of $N+1$ charges q_i, with masses μ_i, where the total mass is M and the total charge is zero. The interaction Hamiltonian is given by

$$H_I = -\sum_{i=0}^{N} \frac{q_i \boldsymbol{A}(r_i) \cdot \boldsymbol{p}_i}{\mu_i}. \tag{13}$$

This has to be rewritten in terms of internal and external variables, by using (A3) from the appendix. The former variables are denoted by Greek symbols, the latter by capital ones. We keep the long-wavelength approximation, but allow the mass M to be finite. We thus include the centre-of-mass motion. Therefore, instead of expanding the field around the origin, we now have an expansion of $\boldsymbol{A}$ around the centre of mass (see appendix),

$$\begin{aligned} \boldsymbol{A}(r_0) &= \boldsymbol{A}(R) - \left(\sum_{i=1}^{N} \frac{\mu_i \rho_i}{M} \cdot \nabla \right) \boldsymbol{A}(R) + \ldots \\ \boldsymbol{A}(r_i) &= \boldsymbol{A}(R) - \left(\sum_{j=1}^{N} \frac{\mu_j \rho_j}{M} \cdot \nabla \right) \boldsymbol{A}(R) + (\rho_i \cdot \nabla) \boldsymbol{A}(R) + \ldots . \end{aligned} \tag{14}$$

Notice that one does not obtain the multipolar Hamiltonian in this way. In order to do so, one should in addition apply a unitary transformation [19, 23]: this step is not necessary, however, for obtaining the selection rules.

The state vector of the atom is given by the direct product of an internal part $|\psi\rangle$ and an external part $|\Psi\rangle$. Now also the latter part can change during the emission or absorption of a photon, leading to selection rules for external variables.

3.2.1. Dipole transitions. To lowest order in a/λ one finds the dipole term

$$H_I^{\mathrm{ED}} = \sum_{i=1}^{N} \left(\frac{q_0}{\mu_0} - \frac{q_i}{\mu_i} \right) \boldsymbol{A}(\boldsymbol{R}) \cdot \boldsymbol{\pi}_i. \tag{15}$$

Note that the *full* spatial dependence of the field has been taken into account, since $\boldsymbol{R}$ is now a dynamical variable. Each single term in the summation leads to the usual dipole selection rules for the corresponding internal variables m_i and l_i, whereas the other quantum numbers for $j \neq i$ do not change. Therefore, for the internal quantum numbers m_i, l_i and also for $m = \sum m_i$ and for $\sum l_i$ one finds the selection rules (7).

Additional selection rules now exist for the external angular momentum. They are found from the requirement that the matrix elements of $\boldsymbol{A}(\boldsymbol{R})$ between the external parts of the atomic initial and final states be non-vanishing. These selection rules depend explicitly on the spatial dependence of the field modes. In particular, they are different for multipole waves and Bessel waves. Explicit examples will be given in section 4.

Hence, the conservation law now refers to the *total* angular momentum, including the external atomic part. This fact implies, for instance, that an electric dipole transition is also possible in a field with angular momentum $j > 1$. This can be understood in two ways. First, the remaining angular momentum can now be absorbed by the external motion of the atom. Second, the field is sampled by the atomic wavefunction, not only at the origin, but around $\boldsymbol{R}$ with a width determined by the de Broglie wavelength. Therefore, the fact that a higher-order multipole field vanishes at the origin, no longer implies that the transition is forbidden.

3.2.2. Higher order transitions. The first-order terms in H_I, which are linear in the gradient of the vector potential, can be grouped according to

$$H_I^1 = \sum_{i=1}^{N} \sum_{j=1}^{N} \left(\frac{q_i}{\mu_i} - \frac{q_0}{\mu_0} \right) \left[\frac{\mu_j \boldsymbol{\rho}_j}{M} \cdot \boldsymbol{\nabla} \right] (\boldsymbol{A}(\boldsymbol{R}) \cdot \boldsymbol{\pi}_i) - \sum_{i=1}^{N} \frac{q_i}{\mu_i} [\boldsymbol{\rho}_i \cdot \boldsymbol{\nabla}](\boldsymbol{A}(\boldsymbol{R}) \cdot \boldsymbol{\pi}_i)$$
$$- \sum_{i=1}^{N} \frac{q_i}{M} [\boldsymbol{\rho}_i \cdot \boldsymbol{\nabla}](\boldsymbol{A}(\boldsymbol{R}) \cdot \boldsymbol{P}). \tag{16}$$

For an atom the ratio μ_i/M is small, so that the first term is negligible compared to the second. The third term describes in effect a dipole coupling of the external motion with the magnetic field: it is equivalent to the Röntgen term, which has recently been shown to be necessary for a consistent description of the centre-of-mass motion for cooled atoms [23], and which leads to the existence of a geometric phase analogous to the Aharonov–Casher effect [24]. We concentrate on the second line, which can be separated into two terms,

$$H_I^{\mathrm{MD}} = \overleftrightarrow{Q}_{\mathrm{MD}}(\boldsymbol{R}) : \sum_{i=1}^{N} \frac{q_i}{\mu_i} (\boldsymbol{\rho}_i \boldsymbol{\pi}_i - \boldsymbol{\pi}_i \boldsymbol{\rho}_i)$$
$$H_I^{\mathrm{EQ}} = \overleftrightarrow{Q}_{\mathrm{EQ}}(\boldsymbol{R}) : \sum_{i=1}^{N} \frac{q_i}{\mu_i} (\boldsymbol{\rho}_i \boldsymbol{\pi}_i + \boldsymbol{\pi}_i \boldsymbol{\rho}_i) \tag{17}$$

where $\overleftrightarrow{Q}_{\mathrm{EQ}}$ and $\overleftrightarrow{Q}_{\mathrm{MD}}$ are given by the same expressions (10) but now evaluated at $\boldsymbol{R}$. For each internal angular momentum quantum number m_i and l_i one finds the same selection

rules (11) and (12) as before. For the external angular momentum one finds additional selection rules from the requirement that the gradient of A have non-zero matrix elements between initial and final external atomic states. Again, the latter rules depend on the explicit spatial dependence of the field modes, and will be different from those in a dipole transition. For details see section 4.

We note that the inclusion of the external atomic motion leads to the following modifications in comparison with the results from the preceding subsection.

(i) In each order of the expansion the atom interacts with the total field.

(ii) In each order the full spatial field dependence is included.

(iii) Each expansion term of the interaction Hamiltonian is invariant under rotations around the origin. Therefore the selection rules express conservation of angular momentum of the total field and the atom.

(iv) External atomic angular momentum is included. The change in internal angular momentum is determined not only by the angular momentum of the field but also by the change in external angular momentum.

4. Illustrations

We start this section by having a closer look at dipole transitions in a field with arbitrary angular momentum $j > 1$. Next we consider electric quadrupole transitions, being the lowest-order transitions in which the atomic angular momentum can change by two units, in a field with angular momentum 1.

4.1. Electric dipole transitions

The transition probability for going from a given initial to some final state in a dipole transition is determined by the square of the matrix element

$$\sum_{i=1}^{N} \langle \Psi_f | \boldsymbol{A}(\boldsymbol{R}) | \Psi_i \rangle \cdot \left(\frac{q_0}{\mu_0} - \frac{q_i}{\mu_i} \right) \langle \psi_f | \pi_i | \psi_i \rangle. \tag{18}$$

If one makes the usual approximation of neglecting the centre-of-mass motion, then first of all the variable $\boldsymbol{R}$ is a fixed position in space. The first part of the matrix element, then, reduces to

$$\boldsymbol{A}(\boldsymbol{R}) \langle \Psi_f | \Psi_i \rangle$$

which implies that, indeed, the external state of the atom does not change. This part will contribute only to the total transition rate, but does not change the angular momentum or the parity of $|\Psi_i\rangle$. Furthermore, if the vector potential vanishes at the centre-of-mass position, then the transition is forbidden. Now, when the external motion is quantized, $\boldsymbol{R}$ becomes a dynamical variable. The matrix element

$$\langle \Psi_f | \boldsymbol{A}(\boldsymbol{R}) | \Psi_i \rangle = \int \mathrm{d}\boldsymbol{R}\, \Psi_f^*(\boldsymbol{R}) \boldsymbol{A}(\boldsymbol{R}) \Psi_i(\boldsymbol{R}) \tag{19}$$

is non-zero only when the external state changes; the external state can change parity and can absorb angular momentum. If $\boldsymbol{A}$ vanishes at the *average* centre-of-mass position, it is no longer implied that the corresponding matrix element vanishes. Still, the electric dipole

transition probability in such a field will be negligibly small in general. For example, in an electric multipole wave with angular momentum j, the field around the origin is proportional to [12, 19]

$$|\boldsymbol{A}(r)| \propto \left(\frac{2\pi r}{\lambda}\right)^{j-1} \tag{20}$$

for $r/\lambda \ll 1$. Thus, if the width of the centre-of-mass wavefunctions of the atom is given by $\Delta x \ll \lambda$, the transition probability is proportional to $(2\pi\Delta x/\lambda)^{2j-2}$, which is negligibly small, except for $j = 1$. For ultracold atoms, however, the width of the wavefunctions of $|\Psi_i\rangle$ and $|\Psi_f\rangle$ is appreciable, as it is determined by the small momentum spread Δp, according to $\Delta x > \hbar/\Delta p$. For atoms cooled down to the recoil limit, $\Delta p \approx h/\lambda$, one obtains

$$\Delta x > \frac{\lambda}{2\pi}. \tag{21}$$

Hence, in this case the field is probed over a distance of the order of the wavelength, so that effectively all higher-order gradients of the field will contribute. Thus the electric dipole transition probability becomes finite, for any multipole wave. As an example, consider an electric quadrupole wave with $j = 2$ and projection $m = 2$. If an atom makes an electric dipole transition, then the selection rules for the internal variables are not changed:

$$\delta l = \pm 1 \qquad \delta m = -1, 0, 1. \tag{22}$$

Thus the atom absorbs only one unit of angular momentum, although the photon contains two units. The deficit is accounted for by the external angular momentum of the atom. For instance, the spherical components of $\boldsymbol{A}$ are proportional to

$$(A_x + \mathrm{i}A_y) \propto \exp(3\mathrm{i}\phi) \qquad A_z \propto \exp(2\mathrm{i}\phi) \qquad (A_x - \mathrm{i}A_y) \propto \exp(\mathrm{i}\phi) \tag{23}$$

so that the selection rules for external quantum number M are

$$\delta M = 3, 2, 1 \tag{24}$$

such that $\delta m + \delta M = 2$. The quantum number J for the length of the total angular momentum $\boldsymbol{J}_{\text{total}} = \boldsymbol{J}_{\text{int}} + \boldsymbol{J}_{\text{ext}}$, changes according to

$$\delta J = \pm 2 \tag{25}$$

which expresses angular momentum conservation. For L, the eigenvalue pertaining to J^2_{ext}, the selection rules are less strict and follow from the familiar triangle and parity rules

$$L + l \geqslant J \geqslant |L - l| \qquad L' + l' \geqslant J' \geqslant |L' - l'| \tag{26}$$

and $\delta L = L' - L$ is odd.

4.2. Electric quadrupole transitions

The transition probability for electric quadrupole transitions is determined by the square of the matrix elements

$$\sum_{i=1}^{N}\langle\Psi_f|\hat{\mathcal{Q}}_{EQ}(\boldsymbol{R})|\Psi_i\rangle : \langle\psi_f|\frac{q_i}{\mu_i}(\rho_i\pi_i+\pi_i\rho_i)|\psi_i\rangle. \tag{27}$$

The main conclusions from the preceding subsection can, *mutatis mutandis*, be carried over to this case. For instance, in a field where the quadrupole tensor vanishes in the origin, the probability of making an electric quadrupole transition is negligble, unless the atom is very cold. As an explicit example we consider a circularly polarized Bessel beam (or a Laguerre–Gaussian beam) propagating in the positive z direction, such that $k_z \approx k$, with $L_z = 0$ and $S_z = \hbar$. From (2) one sees that there is only one non-vanishing component of the field:

$$F_x - iF_y = \sqrt{2}f(k_t, k_z, 0) \tag{28}$$

where $k_t \ll k_z$. Suppose an atom absorbs a photon from this beam by making an electric quadrupole transition. Since only one spherical component (28) of the field is non-vanishing, one is left with a reduced set of quadrupole selection rules

$$\delta m = 0, 1, 2 \tag{29}$$

for the projections of internal angular momentum on the z axis. From the explicit spatial dependence of (28) one gets the following selection rules for the external angular momentum,

$$\delta M = 1, 0, -1 \tag{30}$$

respectively, such that $\delta m + \delta M = 1$. Therefore, the magnetic quantum number m can change by 2 units of angular momentum, even though the field possesses only one unit of spin. The second unit does not come from orbital angular momentum of the photon, since $L_z = 0$. Rather the external motion of the atom loses one unit of angular momentum in that case: $\delta M = -1$. For completeness we note that in a wave with $L_z = m\hbar$ and $S_z = \hbar$, the selection rule for δm is not affected, while for δM one has now

$$\delta M = m + 1, m, m - 1 \tag{31}$$

respectively. The extra amount of orbital angular momentum of the photon is thus always transferred to the centre-of-mass motion of the atom. The same conclusion holds for dipole transitions [17]. Similarly, in an electric dipole wave with $j = 1$, the quadrupole transition matrix elements lead to the rules

$$\delta l = \pm 2 \tag{32}$$

even though the field possesses only one unit angular momentum. Again, the other unit comes from the external motion of the atom, not from orbital angular momentum of light.

5. Discussion and conclusions

If one assumes the centre of mass of an atom to be at rest at a given point, the selection rules for one-photon transitions express angular momentum and parity conservation: as is well known [12], an electric or magnetic 2^j transition with $\delta l = j$ is allowed if and only if the emitted or absorbed photon has total angular momentum j, and parity $(-1)^j$ or $(-1)^{j+1}$, respectively.

When the external motion of the atom is included, things are different: a multipole transition is possible also in fields with lower angular momentum. In particular, we showed that a quadrupole transition with $\delta l = \pm 2$ and $\delta m = \pm 2$ is allowed both in a field with total angular momentum $j = 1$, and in a field with vanishing orbital angular momentum $L_z = 0$, and spin $S_z = \pm 1$. Conversely, a multipole transition is also allowed in a field with higher angular momentum: for instance, an atom may emit a quadrupole photon in a dipole transition. In all these cases there is no violation of conservation of angular momentum or parity, since the external motion can absorb angular momentum and since the parity of the external wavefunction can change sign.

These changes originate from the fact that the external centre-of-mass position $\boldsymbol{R}$ must be considered a dynamical variable. Consequently, both the transition probability and the selection rules depend on the explicit spatial dependence of the field. The internal selection rules, on the other hand, are independent of the mode structure, and do not depend on the inclusion of external atomic motion, quantized or classical.

When is it relevant to include the centre-of-mass motion? If the atom is cooled to low temperatures, then the angular momentum transfer, i.e. the torque, of one photon is not negligible. Thus for a cooled atom one expects to see above-mentioned deviations from the usual emission and absorption behaviour. Indeed, one may note that the de Broglie wavelength of the atom becomes larger as its temperature becomes lower. Therefore the cooled atom sees a larger part of the field around its centre-of-mass position. Hence, the spatial dependence of the field becomes important, and thereby also the orbital angular momentum of the field. This part of the field angular momentum is in general absorbed by the external motion of the atom, in the sense discussed in section 4.

This also indicates how an atom can be made to rotate around a given axis. When the atom is cooled in a Doppler cooling scheme with two counter-propagating Laguerre–Gaussian laser beams with azimuthal index m (i.e. $L_z = m\hbar$ per photon), then at each stimulated absorption the atomic external angular momentum changes by $m\hbar$. The angular momentum will on average not change by spontaneous emissions. The net rate of absorption of external angular momentum is then equal to $\mathrm{d}M/\mathrm{d}t = AP_e m\hbar$, where A is the spontaneous decay rate and P_e the probability for the atom to be in the excited state.

Finally, let us note that we discussed only single-photon transitions. Multi-photon transitions can, to lowest order, be seen as a sequence of electric dipole transitions, and the foregoing conclusions can easily be extended to this case.

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Appendix A. Internal and external variables

We consider an $N+1$-particle system, consisting of particles $i = 0 \ldots N$. Their positions and momenta are denoted by r_i and p_i. Here we will define internal and external position and momentum variables for this system. The internal variables will be denoted by greek symbols, the external variables by capital symbols. The internal position variables ρ_i for $i = 1 \ldots N$ are chosen relative to the position of particle $i = 0$, which for an atom would be the nucleus. The external position vector R is the centre-of-mass position. Thus we define

$$R = \sum_{j=0}^{N} \frac{\mu_j r_j}{M} \qquad \rho_i = r_i - r_0 \tag{A1}$$

for $i = 1 \ldots N$, where $M = \sum \mu_i$ is the total mass. The canonically conjugate momenta follow from these definitions, and are given by

$$P = \sum_{j=0}^{N} p_j \qquad \pi_i = p_i - \frac{\mu_i}{M} \sum_{j=0}^{N} p_j. \tag{A2}$$

These expressions are valid both classically and quantum mechanically, and the quantum operators satisfy the canonical commutation relations. Note that if one defines internal coordinates with respect to the centre of mass, one has an overcomplete set of internal variables. Therefore, these variables have to satisfy a constraint condition, and do not satisfy the canonical commutation rules, which, however, does not lead to any difficulties [23].

The inverse relations read

$$r_0 = R - \sum_{j=1}^{N} \frac{\mu_j \rho_j}{M} \qquad r_i = R + \rho_i - \sum_{j=1}^{N} \frac{\mu_j \rho_j}{M}$$

$$p_0 = \frac{\mu_0}{M} P - \sum_{j=1}^{N} \pi_j \qquad p_i = \frac{\mu_i}{M} P + \pi_i. \tag{A3}$$

Finally, the atomic angular momentum can be separated as

$$J_{\text{atom}} = \sum_{i=0}^{N} r_i \times p_i = R \times P + \sum_{i=1}^{N} \rho_i \times \pi_i \qquad \equiv J_{\text{ext}} + J_{\text{int}}. \tag{A4}$$

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