# Transient optical angular momentum effects in light-matter interactions

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The time evolution of the radiation pressure forces due to the action of laser light on matter in the form of neutral molecules, atoms, and ions is considered when the frequency of the light is comparable to a dipoleallowed transition frequency. We find that the transient regime, applicable from the instant the laser is switched on, is important for the gross motion, provided that the upper-state lifetime  $\Gamma^{-1}$  is relatively long, while the steady-state regime, formally such that  $t \ge \Gamma^{-1}$ , is appropriate for the evaluation of the forces and the dynamics for large  $\Gamma$ . With a focus on the orbital-angular-momentum-endowed laser light, the light-induced timedependent forces and torques are determined and their full time dependence utilized to determine trajectories. Marked differences are found in both translational and rotational features in comparison with the results emerging when the steady-state forces are assumed from the outset. Intricate and detailed atom trajectories are plotted for Laguerre-Gaussian light at near resonance for a transition of Eu<sup>3+</sup> that has a particularly small  $\Gamma$ . The implications of the results for trapping and manipulating atoms and ions using laser light are pointed out and discussed.

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## I. INTRODUCTION

It is well known that the translational motion of atoms and ions as well as molecules (henceforth referred to as "atoms") can be influenced by the radiation pressure forces exerted by laser light [1-3]. If the light beam is endowed with orbital angular momentum, there is, in addition, a light-induced torque which leads to a rotational motion of the atoms about the beam axis [4-8]. The torque influences the rotational motion of matter, both in the bulk and for individual atoms. It arises from the orbital angular momentum content of the light and has been the subject of considerable investigation over the last decade or so [9,10]. The trajectory of a given atom in a given Laguerre-Gaussian light beam will depend on the initial conditions at the instant the beam is switched on. However, most treatments of radiation effects on atoms assume that the steady-state position-dependent forces and associated torques are operative during the entire motion. Evidently, this assumption is not justifiable in general.

We have found that the full time dependence is characterized by a transient regime in which the atoms are subject to forces and torques that differ markedly from their steadystate forms and give rise to new features in the subsequent dynamics, in both the translational and rotational aspects of the motion, effectively modifying the initial conditions long before the steady state is reached.

The investigations of the transient regime and its effects for angular-momentum-endowed laser light require careful derivations of the average forces using density matrix techniques. The forces then enter the equation of motion for the atomic center of mass, leading to the trajectory. In order to demonstrate the importance of the transient regime, we apply the theory to the case of  $Eu^{3+}$  ions in Laguerre-Gaussian light.  $Eu^{3+}$  ions possess a relatively narrow upper-state width for a particular transition, permitting transient effects to manifest themselves in the translational and rotational aspects of the gross motion when the ion is subject to Laguerre-Gaussian light.

This paper is organized as follows. In Sec. II we outline the theoretical framework beginning with a statement of the Hamiltonian appropriate for our model involving an atom of finite mass, with the internal dynamics represented by two quantum levels coupled by interaction with the electromagnetic fields. We then outline the steps and justify the approximations leading to the optical Bloch equations governing the evolution of the state populations in terms of the timedependent density matrix elements for the two-level system. In Sec. III we outline the steps leading to the explicitly timedependent solutions of the optical Bloch equations, known as Torrey's solutions, adapted here to the case of Laguerre-Gaussian light. In Sec. IV we make use of the Torrey solutions to derive the time-dependent spatially varying forces acting on an Eu<sup>3+</sup> ion for three specific cases discussed in Sec. III. We proceed to display the results exhibiting the atomic trajectories for the Eu<sup>3+</sup> ion under the influence of the transient forces of a Laguerre-Gaussian light, comparing the predictions with the situation where steady-state forces are used. Section V analyzes the results and contains further comments and final conclusions.

## **II. FORMALISM**

#### A. Hamiltonian

Our first aim is to outline a derivation leading to the timedependent average force acting on the atom as a mobile center of mass exhibiting gross motion, and the internal dynamics is described by a two-level system interacting with laser light. The total Hamiltonian for the atom plus field can be written as the sum of three terms

$$H = H_F + H_a + H_{int} \tag{1}$$

where  $H_F$  and  $H_a$  are the zero-order Hamiltonians for the laser and the atom, respectively, and are given by

$$H_F = \hbar \omega a^{\dagger} a, \qquad (2)$$

$$H_a = \frac{\mathbf{P}^2}{2M} + \hbar \omega_0 \pi^{\dagger} \pi.$$
(3)

Here  $\pi$  and  $\pi^{\dagger}$  are the ladder operators for the two-level system; **P** is the center-of-mass momentum operator with *M* the mass and  $\omega_0$  the dipole transition frequency. The operators *a* and  $a^{\dagger}$  entering  $H_F$  are the annihilation and creation operators of the laser light and  $\omega$  is its frequency. The last term,  $H_{int}$ , in Eq. (1) is the interaction Hamiltonian coupling the laser light to the two-level system in the electric dipole approximation. Explicitly we have

$$H_{int} = -\boldsymbol{\mu} \cdot \mathbf{E}(\mathbf{R}) \tag{4}$$

where  $\mathbf{E}(\mathbf{R})$  is the electric field evaluated at the center-ofmass position vector  $\mathbf{R}$  and  $\boldsymbol{\mu}$  is the dipole moment vector operator:

$$\boldsymbol{\mu} = \boldsymbol{\mu}_{12}(\boldsymbol{\pi} + \boldsymbol{\pi}^{\dagger}), \tag{5}$$

with  $\mu_{12}$  the transition dipole matrix element.

We are interested in the forces and torque acting on the atom due to a general Laguerre-Gaussian (LG) laser light mode for which the electric field operator can be expressed in quantized form as follows:

$$\mathbf{E}(\mathbf{R}) = i[a\hat{\boldsymbol{\varepsilon}}\mathcal{F}_{klp}(\mathbf{R})e^{i\Theta_{klp}(\mathbf{R})} - \mathbf{H} \cdot \mathbf{c} \cdot ], \qquad (6)$$

where  $\hat{\boldsymbol{\varepsilon}}$  is a mode polarization vector, while  $\mathcal{F}_{klp}(\mathbf{R})$  and  $\Theta_{klp}(\mathbf{R})$  are the mode amplitude function and phase, respectively, given by [11]

$$\mathcal{F}_{klp}(\mathbf{R}) = \mathcal{F}_{k00} \frac{N_{lp}}{(1+z^2/z_R^2)^{1/2}} \left(\frac{\sqrt{2}r}{w(z)}\right)^{|l|} L_p^{|l|} \left(\frac{2r^2}{w^2(z)}\right) e^{-r^2/w^2(z)},$$
(7)

$$\Theta_{klp}(\mathbf{R}) = \frac{kr^2z}{2(z^2 + z_R^2)} + l\phi + (2p + l + 1)\tan^{-1}(z/z_R) + kz.$$
(8)

Here  $\mathcal{F}_{k00}$  may be identified as the amplitude for a plane wave propagating along the *z* axis with wave vector *k*; the coefficient  $N_{lp} = \sqrt{p!/(|l|+p!)}$  is a normalization constant; w(z) is a characteristic width of the beam at axial coordinate *z* and is explicitly given by  $w^2(z) = 2(z^2 + z_R^2)/kz_R$ , where  $z_R$  is the Rayleigh range. The LG mode indices *l* and *p* determine the field intensity distribution and are such that  $l\hbar$  is the orbital angular momentum content carried by each quantum.

In order to arrive at the required expressions for the timedependent forces we need to transform to the interaction picture with respect to the zero-order field Hamiltonian  $H_F$ = $\hbar \omega a^{\dagger} a$ . The operators *a* and  $a^{\dagger}$  are then time dependent such that

$$a(t) = e^{iH_F t/\hbar} a e^{-iH_F t/\hbar} = a e^{-i\omega t}.$$
(9)

In the classical limit, appropriate for the case of a coherent beam, the *a* and  $a^{\dagger}$  operators behave as

$$a(t) \to \alpha e^{-i\omega t}, \quad a^{\dagger}(t) \to \alpha^* e^{i\omega t}.$$
 (10)

Substituting for the field operator from Eq. (6), and applying the rotating-wave approximation, we have for the interaction Hamiltonian

$$H_{int} = -\boldsymbol{\mu} \cdot \mathbf{E}(\mathbf{R}) = -i\hbar [\,\tilde{\boldsymbol{\pi}}^{\dagger} f(\mathbf{R}) - \mathbf{H} \cdot \mathbf{c} \,.\,], \qquad (11)$$

where we have introduced  $\tilde{\pi}$  and  $f(\mathbf{R})$  as follows:

$$\widetilde{\boldsymbol{\pi}} = \boldsymbol{\pi} e^{i\omega t}, \quad f(\mathbf{R}) = (\boldsymbol{\mu}_{12} \cdot \widehat{\boldsymbol{\epsilon}}) \, \boldsymbol{\alpha} \mathcal{F}(\mathbf{R}) e^{i\Theta(\mathbf{R})} / \hbar, \qquad (12)$$

and we have, for the time being, dropped the labels klp in the field amplitude function  $\mathcal{F}_{klp}(\mathbf{R})$  and phase  $\Theta_{klp}(\mathbf{R})$ . It is also convenient at this stage to introduce the position-dependent Rabi frequency  $\Omega(\mathbf{R})$  as follows

$$\hbar\Omega(\mathbf{R}) = |(\boldsymbol{\mu}_{12} \cdot \hat{\boldsymbol{\epsilon}}) \alpha \mathcal{F}(\mathbf{R})|, \quad f(\mathbf{R}) = \Omega(\mathbf{R})e^{i\Theta(\mathbf{R})}. \quad (13)$$

Clearly  $\Omega$  also depends on the mode type and would normally bear the labels *klp*.

### **B.** Optical Bloch equations

In order to set up the appropriate optical Bloch equations for the atomic density matrix elements we now allow the position operator **R** and the momentum operator **P** to be replaced by their average values **r** and  $P_0=MV$ , where **V** is the center-of-mass velocity. This semiclassical approximation amounts to treating the center-of-mass motion of the atom classically, while the internal dynamics continues to be treated quantum mechanically. The validity of this approximation demands that the spread in the atomic wave packet be much smaller than the wavelength of the light, and that the recoil energy be much smaller than the linewidth.

Within the semiclassical approximation, the system density matrix associated with the two levels and the center of mass can be written as

$$\rho_{S} = \delta(\mathbf{R} - \mathbf{r}) \,\delta(\mathbf{P} - M\mathbf{V}) \rho(t). \tag{14}$$

The time evolution of the internal density matrix  $\rho(t)$  is such that

$$\frac{d\rho}{dt} = -\frac{i}{\hbar} [H,\rho] + \mathcal{R}\rho, \qquad (15)$$

where the term  $\mathcal{R}\rho$  incorporates the relaxation effects in the two-level system.

The optical Bloch equations for the two-level density matrix elements emerge from the above formalism in the following matrix form:

$$\begin{bmatrix} \dot{\rho}_{21}(t) \\ \dot{\rho}_{12}(t) \\ \dot{\rho}_{22}(t) \end{bmatrix} = \begin{bmatrix} -(\Gamma_2 - i\Delta) & 0 & 2f(\mathbf{r}) \\ 0 & -(\Gamma_2 + i\Delta) & 2f^*(\mathbf{r}) \\ -f^*(\mathbf{r}) & -f(\mathbf{r}) & -\Gamma_1 \end{bmatrix} \begin{bmatrix} \hat{\rho}_{21}(t) \\ \hat{\rho}_{12}(t) \\ \rho_{22}(t) \end{bmatrix} + \begin{bmatrix} -f(\mathbf{r}) \\ -f^*(\mathbf{r}) \\ 0 \end{bmatrix}$$
(16)

where the spontaneous emission process has been described

in terms of an inelastic collision rate  $\Gamma_1$  and an elastic collision one  $\Gamma_2$ . Elastic collisions often have dominant linebroadening effects for a wide variety of physical conditions [12]. We have also introduced the effective, velocitydependent, detuning  $\Delta$  by  $\Delta = \Delta_0 - \nabla \Theta \cdot \mathbf{V}$  (where  $\Delta_0 = \omega - \omega_0$ ) that arises naturally in the expression for the density matrix, and set  $\hat{\rho} = \tilde{\rho} \exp(-it\mathbf{V} \cdot \nabla \Theta)$ . The matrix element  $\rho_{11}$ has been written in terms of  $\rho_{22}$  since  $\rho_{11}(t) + \rho_{22}(t) = 1$ .

## **III. TORREY'S SOLUTIONS**

We use Laplace transform techniques to obtain timedependent solutions to the optical Bloch equations as first employed by Torrey to solve nuclear Bloch equations [13,14]. The Laplace transform is

$$\overline{F}(\lambda) = \int_0^\infty F(t)e^{-\lambda t}dt.$$
 (17)

Applying this transform to the optical Bloch equations leads to the new Laplace transformed optical Bloch equations, expressed here in matrix form,

$$\Lambda \begin{bmatrix} \bar{\rho}_{21}(\lambda) \\ \bar{\rho}_{12}(\lambda) \\ \bar{\rho}_{22}(\lambda) \end{bmatrix} = \begin{bmatrix} -f(\mathbf{r})/\lambda \\ -f^*(\mathbf{r})/\lambda \\ 0 \end{bmatrix},$$
(18)

where the initial conditions are  $\bar{\rho}_{21}(t=0)=\bar{\rho}_{12}(t=0)=\bar{\rho}_{22}(t=0)=0$ , and

$$\mathbf{\Lambda} = \begin{bmatrix} \lambda + \Gamma_2 - i\Delta & 0 & -2f(\mathbf{r}) \\ 0 & \lambda + \Gamma_2 + i\Delta & -2f^*(\mathbf{r}) \\ f^*(\mathbf{r}) & f(\mathbf{r}) & \lambda + \Gamma_1 \end{bmatrix}.$$
 (19)

From Eq. (18) we find

$$\lambda |\Lambda| \overline{\rho}_{21}(\lambda) = -f(\mathbf{r})(\lambda + \Gamma_2 + i\Delta)(\lambda + \Gamma_1),$$
  

$$\lambda |\Lambda| \overline{\rho}_{12}(\lambda) = -f^*(\mathbf{r})(\lambda + \Gamma_2 - i\Delta)(\lambda + \Gamma_1),$$
  

$$\lambda |\Lambda| \overline{\rho}_{22}(\lambda) = 2|f(\mathbf{r})|^2(\lambda + \Gamma_2).$$
(20)

Each equation in Eqs. (20) can be written in the same general form. Assuming that  $\overline{P}$  represents either  $\overline{\rho}_{21}$ ,  $\overline{\rho}_{12}$ , or  $\overline{\rho}_{22}$ , then Eqs. (20) have the form

$$\overline{P}(\lambda) = \frac{g(\lambda)}{\lambda |\Lambda|},\tag{21}$$

where  $g(\lambda)$  represents the corresponding right-hand side of Eq. (20).

If we assume that  $|\Lambda|(\lambda)$  can be factorized into the form

$$|\mathbf{\Lambda}|(\mathbf{\lambda}) = (\mathbf{\lambda} + a)[(\mathbf{\lambda} + b)^2 + s^2]$$
(22)

then  $\overline{P}(\lambda)$  can be written as partial fractions,

$$\overline{P}(\lambda) = \frac{A}{\lambda + a} + \frac{B(\lambda + b) + C}{(\lambda + b)^2 + s^2} + \frac{D}{\lambda}.$$
 (23)

Inverting the transform of this equation leads to solutions to the optical Bloch equations that take the form

$$P(t) = Ae^{-at} + \left(B\cos(st) + \frac{C}{s}\sin(st)\right)e^{-bt} + D.$$
(24)

Such solutions are known as Torrey solutions, in which the first three terms represent transient effects, and the final term the steady state. The coefficients A and D are obtained by taking the following limits:

$$A = \lim_{\lambda \to -a} [(\lambda + a)P(\lambda)],$$
$$D = \lim_{\lambda \to 0} [\lambda \overline{P}(\lambda)],$$

while the coefficients *B* and *C* are found using the relevant initial conditions.

As the aim is to seek time-dependent solutions to the optical Bloch equations in order to find an expression for the transient force exerted on a two-level atom by a Laguerre-Gaussian beam, we need only concentrate on  $\bar{\rho}_{21}$ . This is the only time-dependent factor entering the force expression  $\langle \mathbf{F}(t) \rangle = i\hbar \nabla [\tilde{\rho}_{21}^* f(\mathbf{r}) - \tilde{\rho}_{21} f^*(\mathbf{r})]$ . For  $\bar{\rho}_{21}$  we obtain

$$A = \frac{-g(-a)}{a[(b-a)^2 + s^2]},$$
(25)

$$D = \frac{g(0)}{a(b^2 + s^2)},$$
 (26)

and

$$B = -(A+D), \tag{27}$$

$$C = aA + bB - f(\mathbf{r}). \tag{28}$$

The Torrey rate parameters *a*, *b*, and *s* can be determined by solving the cubic equation  $|\Lambda|=0$ . We consider three special cases of physical interest in which the cubic equation has simple roots. These cases are those of exact resonance, strong collisions, and intense external field.

#### A. Exact resonance

In the case of exact resonance we take the total detuning  $\Delta$  to be zero. The cubic equation becomes

$$|\mathbf{\Lambda}| = (\mathbf{\lambda} + \Gamma_2)[(\mathbf{\lambda} + \Gamma_2)(\mathbf{\lambda} + \Gamma_1) + 4|f(\mathbf{r})|^2] = 0$$
(29)

and the roots are

$$\lambda = -\Gamma_2, \quad -\frac{1}{2}(\Gamma_1 + \Gamma_2) \pm i\sqrt{4|f(\mathbf{r})|^2 - \frac{1}{4}(\Gamma_1 - \Gamma_2)^2}.$$
(30)

This is consistent with rate parameters a, b, and s in the Torrey solution Eq. (24), given by

$$b = \frac{1}{2}(\Gamma_1 + \Gamma_2)$$

 $a - \Gamma_{a}$ 

and with

$$A = 0$$
,

$$B = \frac{f(\mathbf{r})\Gamma_1}{\Gamma_1\Gamma_2 + 4|f(\mathbf{r})|^2},$$

$$C = \frac{f(\mathbf{r})(\Gamma_1^2 - \Gamma_1\Gamma_2 - 8|f(\mathbf{r})|^2)}{2(\Gamma_1\Gamma_2 + 4|f(\mathbf{r})|^2)},$$

$$D = \frac{-f(\mathbf{r})\Gamma_1}{\Gamma_1\Gamma_2 + 4|f(\mathbf{r})|^2}.$$
(32)

#### **B. Strong collisions**

For the case of strong collisions we have a large number of atoms in the system. Thus, we take the elastic and inelastic collision rates to be approximately equal and we can therefore write

$$\Gamma \equiv \Gamma_1 \approx \Gamma_2. \tag{33}$$

Therefore, the cubic equation becomes

$$|\mathbf{\Lambda}| = (\mathbf{\lambda} + \Gamma)[(\mathbf{\lambda} + \Gamma)^2 + 4|f(\mathbf{r})|^2] + \Delta^2(\mathbf{\lambda} + \Gamma) = 0 \quad (34)$$

with roots

$$\lambda = -\Gamma, \quad -\Gamma \pm \sqrt{\Delta^2 + 4|f(\mathbf{r})|^2}. \tag{35}$$

This implies that the rate parameters a, b, and s in the Torrey solution Eq. (24) are

$$a = b = \Gamma$$
,

$$s = \sqrt{\Delta^2 + 4|f(\mathbf{r})|^2} \tag{36}$$

$$A = 0,$$
  

$$B = \frac{f(\mathbf{r})(\Gamma + i\Delta)}{\Gamma^2 + \Delta^2 + 4|f(\mathbf{r})|^2},$$
  

$$C = \frac{f(\mathbf{r})[i\Delta\Gamma - \Delta^2 - 4|f(\mathbf{r})|^2]}{[\Gamma^2 + \Delta^2 + 4|f(\mathbf{r})|^2]},$$
  

$$D = \frac{-f(\mathbf{r})(\Gamma + i\Delta)}{\Gamma^2 + \Delta^2 + 4|f(\mathbf{r})|^2}.$$
(37)

#### C. Intense external field

Finally we consider the case where the intensity of the light beam is sufficiently large that  $\Omega/\Gamma_2 \gg 1$ , in which case  $\Omega/\Gamma_1 \gg 1$  is also true. Therefore,

$$\Omega \gg \gamma \tag{38}$$

where  $\gamma \equiv \Gamma_2 - \Gamma_1$ . We now rewrite  $|\Lambda|$  in terms of  $\Gamma_2$  and  $\gamma$ , and we have

$$|\mathbf{\Lambda}| = (\mathbf{\lambda} + \Gamma_2)[(\mathbf{\lambda} + \Gamma_2)(\mathbf{\lambda} + \Gamma_2 - \gamma) + 4|f(\mathbf{r})|^2] + \Delta^2(\mathbf{\lambda} + \Gamma_2 - \gamma) = 0$$
(39)

or

with

$$(\lambda + \Gamma_2)[(\lambda + \Gamma_2)^2 + 4|f(\mathbf{r})|^2 - \gamma(\lambda + \Gamma_2) + \Delta^2] = \gamma \Delta^2.$$
(40)

When the Rabi frequency  $\Omega$  is very large compared to  $\gamma$ , Eq. (40) has three roots which can be found as follows. First, if we assume that  $(\lambda + \Gamma_2)^2 \ll \Delta^2 + 4|f(\mathbf{r})|^2$ , we can rearrange Eq. (40) to the form

$$(\lambda + \Gamma_2) = \frac{\gamma \Delta^2}{[\Delta^2 + 4|f(\mathbf{r})|^2]\{1 + (\lambda + \Gamma_2)(\lambda + \Gamma_2 - \gamma)/[\Delta^2 + 4|f(\mathbf{r})|^2]\}}.$$
(41)

This leads to the first root

$$\lambda \approx -\Gamma_2 + \gamma \frac{\Delta^2}{\Delta^2 + 4|f(\mathbf{r})|^2} + O\left(\left(\frac{\gamma}{\Omega}\right)^3\right). \tag{42}$$

To obtain the second and third roots we consider the case where  $(\lambda + \Gamma_2)^2 \approx [\Delta^2 + 4|f(\mathbf{r})|^2]$ , leading to an alternative rearrangement of Eq. (40) of the form

$$(\lambda + \Gamma_2)^2 + \Delta^2 + 4|f(\mathbf{r})|^2 = \gamma(\lambda + \Gamma_2) \left(1 + \frac{\Delta^2}{(\lambda + \Gamma_2)^2}\right),$$
(43)

which yields the second and third roots

$$\lambda = -\Gamma_2 + \frac{2\gamma |f(\mathbf{r})|^2}{\Delta^2 + 4|f(\mathbf{r})|^2} \pm i\sqrt{\Delta^2 + 4|f(\mathbf{r})|^2} + O\left(\left(\frac{\gamma}{\Omega}\right)^2\right).$$
(44)

Thus, the Torrey rate parameters a, b, and s in the intense external field case are

$$a = \Gamma_2 - (\Gamma_2 - \Gamma_1) \frac{\Delta^2}{\Delta^2 + 4|f(\mathbf{r})|^2},$$
  

$$b = \Gamma_2 - \frac{2(\Gamma_2 - \Gamma_1)|f(\mathbf{r})|^2}{\Delta^2 + 4|f(\mathbf{r})|^2},$$
  

$$s = \sqrt{\Delta^2 + 4|f(\mathbf{r})|^2}.$$
(45)

In this case we find that the Torrey rate parameters have a more complicated form than in the other two cases, and it is therefore more convenient to leave the coefficients A, B, C, and D written in terms of them. Hence, the coefficients in the case of an intense external field are expressible as

$$A = \frac{f(\mathbf{r})(\Gamma_2 + i\Delta - a)(\Gamma_1 - a)}{a[(b-a)^2 + s^2]},$$
  

$$B = -(A+D),$$
  

$$C = aA + bB - f(\mathbf{r}),$$
  

$$D = -\frac{f(\mathbf{r})\Gamma_1(\Gamma_2 + i\Delta)}{a(b^2 + s^2)}.$$
(46)

Having established the time dependence for the density matrix elements, for the special cases described, we are now in a position to derive the time-dependent forces acting on a  $Eu^{3+}$  ion subject to a Laguerre-Gaussian beam.

# IV. DYNAMICS OF Eu<sup>3+</sup> ION IN LG LIGHT

The average force exerted by the light on the ion center of mass is the expectation value of the rate of change of the center-of-mass momentum, which amounts to the expectation value of the trace of  $-\rho \nabla H_{int}$ ,

$$\langle \mathbf{F} \rangle = - \langle \operatorname{tr}(\rho \, \nabla \, H_{int}) \rangle.$$
 (47)

This is a time-dependent as well as spatially dependent force and it turns out that it is divisible into two types of force, namely, the dissipative force  $\langle \mathbf{F}_{diss} \rangle$  and a dipole force  $\langle \mathbf{F}_{dipole} \rangle$ . It can be shown that these forces are related to the density matrix elements by the expressions [5]

$$\langle \mathbf{F}_{diss}(\mathbf{R}, \mathbf{t}) \rangle = -\hbar \, \boldsymbol{\nabla} \, \Theta[\hat{\rho}_{21}^* f(\mathbf{r}) + \hat{\rho}_{21} f^*(\mathbf{r})], \qquad (48)$$

$$\langle \mathbf{F}_{dipole}(\mathbf{R}, \mathbf{t}) \rangle = i\hbar \frac{\nabla \Omega}{\Omega} [\hat{\rho}_{21}^* f(\mathbf{r}) - \hat{\rho}_{21} f^*(\mathbf{r})].$$
(49)

In order to explore the transient regime we consider the dynamics of an atom in the field of a Laguerre-Gaussian beam from the instant the beam has been switched on. The relevant equation determining the dynamics is Newton's second law, written in the form

$$M\frac{d^2\mathbf{R}}{dt} = \langle \mathbf{F}(t) \rangle \tag{50}$$

subject to initial conditions. Since the equation of motion is second order in time we need two sets of initial conditions,

namely, values of the initial position vector components  $\mathbf{R}(0)$ and values of the initial velocity vector components  $\mathbf{V}(0)$ .

The main by-product of solving the equation of motion is the trajectory  $\mathbf{R}(t)$ , but the solutions should also determine the evolution of velocity and acceleration. The associated evolution of the forces also enables determination of the torque acting on the center of mass, responsible for rotational motion.

In previous investigations on atom dynamics in LG beams, a magnesium ion was considered in the study of the effects of the steady-state forces arising from a two-level atom in angular-momentum-endowed light [5]. This is not a suitable choice for considering transient effects as, due to the short lifetime of the excited state of the magnesium ion, the steady-state forces will dominate, making the transient forces negligible. To illustrate transient effects we need to consider transitions with a long upper-state lifetime, and rare-earth ions are good examples of this. In particular, we consider a Eu<sup>3+</sup> ion in the numerical work that now follows [15].

The atomic mass of Eu<sup>3+</sup> is  $25.17 \times 10^{-26}$  kg and the relevant transition is  ${}^{5}D_{0} \rightarrow {}^{7}D_{1}$ , which corresponds to a wavelength  $\lambda = 614$  nm. The upper state for this transition has  $\Gamma$ =1111.11 Hz. We concentrate on the l=1, p=0 Laguerre-Gaussian mode. We take the laser intensity to be I=10<sup>5</sup> W cm<sup>-2</sup>, except for the intense external field special case where the intensity is  $I=10^{8}$  W cm<sup>-2</sup>. The beam waist is taken to be  $w_{0}=35\lambda$ .

All the numerical evaluations considered here were run for a period  $t_{\text{max}} \approx 5\Gamma^{-1}$ , which is approximately 4.5 ms, allowing for the transient regime to manifest itself in the results and for the steady-state forces to be the appropriate forces at the end of the run.

### A. Exact resonance

On the assumption that  $\Gamma_2 = \Gamma_1/2 = \Gamma$ , then in the case of exact resonance the dissipative force emerges from the procedure described above in the form

$$\langle \mathbf{F}_{diss} \rangle = \frac{-\hbar\Omega^2 \Gamma \, \nabla \, \Theta}{\Gamma^2 + 2\Omega^2} \Biggl\{ \Biggl[ \cos(st) \\ - \left( \frac{\Gamma}{2} + \frac{4\Omega^2}{\Gamma} \right) \frac{\sin(st)}{s} \Biggr] e^{-(3/2)\Gamma t} - 1 \Biggr\}$$
(51)

where  $s = \sqrt{4\Omega^2 - \frac{1}{4}\Gamma^2}$ . There is no dipole force acting on the atom in the case of exact resonance, which also means that there is no radial force acting on the atom (in cylindrical coordinates). Therefore, if the atom is initially at rest at a point off the beam axis, the atom will remain at the same radial distance from the beam axis throughout the trajectory. This can be seen in Fig. 1, where the Eu<sup>3+</sup> ion starts from rest in the *x*-*y* plane at distance  $\lambda$  from the beam axis and rotates around the axis, traveling forward in the direction of the beam propagation. This is the expected behavior in which the atom experiences no evidence of trapping under exact resonance conditions.



FIG. 1. Trajectory of a  $Eu^{3+}$  ion starting at rest in a  $LG_{1,0}$  mode in the case of exact resonance. The dot represents the initial position of the atom. See the text for values of the other parameters needed for the evaluation of the trajectory.

#### **B. Strong collisions**

For strong collisions, the dissipative force emerges in the form

$$\langle \mathbf{F}_{diss} \rangle = \frac{-2\hbar\Omega^{2}\Gamma\,\boldsymbol{\nabla}\,\Theta}{\Delta^{2} + \Gamma^{2} + 4\Omega^{2}} \\ \times \left\{ \left[ \cos(st) - \left(\frac{\Delta^{2} + 4\Omega^{2}}{\Gamma}\right)\frac{\sin(st)}{s} \right] e^{-\Gamma t} - 1 \right\}$$
(52)

and the corresponding dipole force is

$$\langle \mathbf{F}_{dipole} \rangle = \frac{2\hbar\Delta\Omega\,\nabla\,\Omega}{\Delta^2 + \Gamma^2 + 4\Omega^2} \left[ \left(\cos(st) - \frac{\Gamma\,\sin(st)}{s\Delta}\right) e^{-\Gamma t} - 1 \right]$$
(53)

where  $\Gamma \equiv \Gamma_2$  and  $s = \sqrt{\Delta^2 + 4\Omega^2}$ . The evolution of the dynamical state of the Eu<sup>3+</sup> motion in a Laguerre-Gaussian beam can be followed for a relatively long period of the order  $5\Gamma^{-1}$ . Figure 2 displays the trajectory of the ion in the case of strong collisions with initial position on the *z* plane at radial coordinates  $r=5\lambda$  in Fig. 2.

#### C. Intense external field

For an intense external field the expressions for the transient forces obtained are more complex than for the previous two cases. It is convenient to introduce the parameters  $\chi$  and  $\zeta$  related to the Torrey rate parameters through  $\chi = (\Gamma -2a)/2a[(b-a)^2+s^2]$  and  $\zeta = \Gamma/2a(b^2+s^2)$ . In terms of  $\chi$  and  $\zeta$  the dissipative force emerges in the form

$$\langle \mathbf{F}_{diss} \rangle = -2\hbar\Omega^2 \, \boldsymbol{\nabla} \, \Theta \Bigg[ (\Gamma - a)\chi e^{-at} + \left( [\Gamma\zeta - (\Gamma - a)\chi] \cos(st) + \{a(\Gamma - a)\chi - b[(\Gamma - a)\chi - \Gamma\zeta] - 1\} \frac{\sin(st)}{s} \right) e^{-bt} - \Gamma\zeta \Bigg]$$

$$(54)$$

while the dipole force is



FIG. 2. Trajectory of a Eu<sup>3+</sup> ion projected in the *x*-*y* plane with the ion starting at rest in a  $LG_{1,0}$  mode in the case of strong collisions. The dot represents the initial position of the ion. See the text for values of parameters used in the generation of this trajectory

$$\langle \mathbf{F}_{dipole} \rangle = 2\hbar \Delta \Omega \, \boldsymbol{\nabla} \, \Omega \bigg[ \chi e^{-at} + \left( (\zeta - \chi) \cos(st) + \left[ a\chi - b(\chi - \zeta) \right] \frac{\sin(st)}{s} \right) e^{-bt} - \zeta \bigg].$$
 (55)

The parameters *a*, *b*, and *s* are given in Eq. (45). We have also written  $\Gamma \equiv \Gamma_2 \approx \frac{1}{2}\Gamma_1$  as was assumed in the exact resonance case. Figure 3 displays the trajectory for a Eu<sup>3+</sup> ion subject to the Laguerre-Gaussian beam in the intense external field case where the atom is initially at rest in the *z*=0 plane at a radial coordinate *r*=5 $\lambda$ . The total duration of the trajectory is 5 $\Gamma^{-1}$ , and the detuning is  $\Delta_0$ =100 $\Gamma$ .

#### V. COMMENTS AND CONCLUSIONS

In the cases of strong collisions and intense external field the atomic trajectory within the body of the Laguerre-



FIG. 3. As in Fig. 2, but here the case is that of strong external field. The dot represents the initial position of the ion. See the text for values of parameters used in the generation of this trajectory



FIG. 4. The portion of the projected trajectory in Fig. 1 showing how the loops are formed. The inset to the figure shows the evolution of the x coordinate with time.

Gaussian beam follows a characteristic path in which there is axial motion superimposed on an in-plane motion. The inplane trajectory is in the form of loops resembling the petals of a flower. The manner in which the trajectory takes shape can be seen with reference to Fig. 4 which displays a portion of the trajectory exhibited for the case considered in Fig. 2. We see from the main Fig. 4 that as the atom approaches the beam axis (where there is zero intensity), it first travels across and experiences a change in the sign of the torque after crossing the axis. It then follows a curved downward path before it gets repelled at some radial position determined by the dipole potential profile. It then turns upwards toward the axis, and completes the first loop by crossing the axis again, but this time it approaches from below, so it experiences another change in the sign of the torque. It then follows a similar pattern, completing a second upper loop. In this manner the first two loops form the shape of the number 8. The third loop is displaced azimuthally relative to the first, and so on. The inset to the figure confirms the order in which the first and second loops are formed, whereby the x coordi-



FIG. 5. The evolution of the torque being exerted on the Eu<sup>3+</sup> ion in the strong collisions case (with  $\Delta_0=500$ ), when the initial position of the atom is  $r=5\lambda$ .



FIG. 6. Variation with time of the steady-state time-independent torque exerted on Eu<sup>3+</sup>: exact resonance (full curve), strong collisions (dotted curve), and intense external field (dot-dashed curve).

nates after the first crossing of the axis conform with the formation of the lower loop first. It is in the creation of the petal-like trajectory and in the azimuthal displacement of the loops that the influence of the light-induced torque manifests itself. After a sufficiently long time, the forces and torque approach steady-state values, corresponding to the timeindependent (i.e., post-transient) parts of the force expressions in Sec. III.



FIG. 7. Trajectory of a Eu<sup>3+</sup> ion in a LG<sub>1,0</sub> beam initially at rest  $5\lambda$  from the beam axis in the steady-state regime using the parameters for the special cases of (a) strong collisions and (b) intense external field. The initial position of the atom is represented by the dot.

	$r_0/\lambda$	$\Delta_0/\Gamma$	$I ({\rm W}{\rm cm}^{-2})$	$\langle r_f \rangle / \lambda$	$ \langle T \rangle /\hbar k \Gamma \lambda$
Exact resonance	1	0	10 <sup>5</sup>	1.000	1.000
Strong collisions	1	500	10 <sup>5</sup>	21.212	0.503
	5	500	10 <sup>5</sup>	1.027	0.268
Intense external field	5	100	10 <sup>8</sup>	2.314	0.945

TABLE I. The average torque exerted on the atom in the steady-state régime  $(5 < \Gamma t < 10)$  for three special cases, where  $\langle r_f \rangle$  is the average radial position from the beam axis during the steady state.

A similar behavior is exhibited in the case of an intense external field, Fig. 3. However, the number of the loops is greatly increased; this is clearly due to the increase in kinetic energy of the atom, both translational (radially due to stronger radial forces) as well as rotationally (due to the torque) in the presence of the more intense field of the Laguerre-Gaussian beam.

The looped trajectory cannot arise in the case of exact resonance, Fig. 1, since there is no dipole force acting on the atom and hence no radial force, due to the zero detuning. The radial position remains constant, as is indeed exhibited in the figure.

It is instructive to examine the evolution of the torque corresponding to the trajectories in Figs. 2 and 3. Figure 5 shows the torque experienced by the Eu<sup>3+</sup> ion in the case of strong collisions when the atom has an initial position of  $r = 5\lambda$  from the beam axis, and the detuning is 500 $\Gamma$ . The formal expression for the time-dependent torque is

$$\mathbf{\mathcal{T}}(t) = \mathbf{r}(t) \times \langle \mathbf{F}(t) \rangle.$$
(57)

Note that the time evolution of the torque is due to the fact that both the position and the average force carry time dependence. From Fig. 5 it is easy to see that immediately after the Laguerre-Gaussian beam is switched on, there is a sudden increase in the torque, which subsequently oscillates between positive and negative values. This large torque quickly decays away toward a steady-state torque corresponding to the time-independent part of the dissipative force. Furthermore, it can be seen that, after the initial response, a collapse and revival pattern is exhibited by the evolution of the torque. We have checked by explicit analysis that this behavior corresponds to the individual loops in the trajectory, with the peak of the torque corresponding to the outer tip of the loop, and the collapse of the torque to the points in the trajectory where the atom is close to the centre of the beam. We have also checked that the sudden jump discontinuities in the torque are real events arising from the change in the direction of motion as the atom crosses the axis.

It is reassuring to check whether our procedure, which employs the full time dependence of the forces to determine the dynamics, does indeed correctly represent the large-time limit. Table I shows the mean average of the torque on the atom during the long-time interval  $5 < \Gamma t < 10$  for the special cases of exact resonance, strong collisions, and intense external field. The average radial position from the beam axis during the same period was also recorded so that the values of the average torque can be compared with the time-independent torque, arising from the steady-state term of the Torrey solutions.

The formal expressions for the steady-state forces can be deduced from Eqs. (16) by taking the limit  $t \rightarrow \infty$ . The variation of the torques, formally given by Eq. (57) with radial position are shown in Fig. 6 for the three cases discussed. It is easy to check that the results in Table I are consistent with those in Fig. 6. Finally we present the results of the dynamics in which the steady-state forces are assumed from the outset. Figure 7 shows the trajectory for the same situations as in Figs. 2 and 3. The differences are clear and we conclude that steady-state forces do not correctly describe the motion of atoms subject to Laguerre-Gaussian beams for transitions possessing long upper-state lifetimes.

In conclusion, we have shown that transient effects due to the interaction of atoms with a Laguerre-Gaussian beam become significant when the upper-state lifetime is relatively long. The evolutions of the dynamical variables of the gross motion are markedly different from those arising from the situation determined by assuming the validity of the steadystate forces from the outset. The trajectories determined here suggest that under certain circumstances consideration of the transient regime is crucial for the correct dynamics. It should always be taken into consideration in the processes of trapping and, in general, manipulating atomic beams using laser light for transitions possessing a long lifetime for the upper state.

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