Theory of radiation forces and momenta for mobile atoms in light fields

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(Received 18 February 1993)

A theory for a bound system of charges interacting with electromagnetic fields is developed, with special emphasis on the radiation-pressure effects imposed by the fields on the gross motion of the charges. The theory is particularly simple and transparent for the case of two opposite charges of finite mass M in the electric-dipole approximation. This enables a rigorous description to be given for the quantum electrodynamics of the problem with the various interaction terms affecting the gross motion being easily identified. Here, too, it becomes clear that, even in the dipole approximation, the conventional interaction term should be supplemented by the so-called Röntgen term. Besides ensuring the consistency of the theory for the overall (charges plus fields) system regarding conservation laws, the Röntgen term has dynamical consequences. This is established by explicit calculations of the expectation values of the mechanical momentum $\langle M\dot{\mathbf{R}} \rangle$ and the radiation pressure force $\langle M\ddot{\mathbf{R}} \rangle$ on a two-level atom in the dipole approximation. Results of calculations are displayed for the case of a single light beam and for counterpropagating beams. The feasibility of experimentally observing the transverse force is explored. Generalizations of the well-known friction force arising in one-dimensional optical molasses are given and discussed.

PACS number(s): 42.50.Vk

I. INTRODUCTION

The advent of tunable lasers has led to the detection of previously undetected phenomena in which the atomic gross motion is the main ingredient [1]. A well-known model for exploring the basic physics in this context involves the interaction between laser light and a mobile two-level atom. This simple model has been very useful in providing insight into the radiation pressure force and the corresponding momentum exchange between light and matter in a number of physical situations—for example, in atomic beam deflection and focusing [2], and for elucidating the so-called Doppler mechanism in laser cooling [3]. It is also useful for studying the collective properties of laser-cooled trapped atoms and ions [4].

In a recent paper [5], a general theory was presented dealing with an ensemble of an arbitrary number of charges forming a bound system interacting with transverse electromagnetic fields. One of the main features of the results emerging from the theory was the need to distinguish between the canonical and mechanical momenta of the gross motion of the ensemble and, in so doing, the formalism automatically led to the prediction of additional interaction terms involving a coupling of the fields to the Röntgen current.

The purpose of the present paper is twofold. First, we show that similar conclusions as regards the presence of the Röntgen contribution are obtained when we consider a system of two bound charges of finite total mass M. This simplifies considerably the formalism needed for effecting the division of the motion into an internal plus a gross motion, which also involves application of a unitary transformation on the corresponding Hamiltonian. Secondly, we apply the formalism, including the additional interaction, to the calculation of specific dynamical attributes of the gross motion, namely the time evolution of the expectation values of the mechanical momentum and of the pressure force due to irradiation. The method is the only canonical, and therefore rigorous, procedure that allows for the introduction into the formalism of velocity-dependent terms. As will be shown, this treatment enables a number of interesting special cases to be considered.

The plan of this paper is as follows. In Sec. II we consider the system of two charges as a prototype atomic system interacting with light. We follow the usual procedure leading from the conventional Lagrangian in the Coulomb (radiation) gauge to the Hamiltonian, identifying the canonical variables and corresponding momenta. On the Hamiltonian we perform a unitary transformation which has the advantage of facilitating the division between the center of mass and internal variables. The nonrelativistic Hamiltonian is then derived, which is valid in the electric-dipole approximation, and in which gross motion attributes are explicit. The additional interaction term, in the dipole approximation, is pointed out and shown to involve a coupling between the field and the internal and gross motions of the atomic system. In Secs. III and IV we apply the formalism including the new interaction terms to explore the dynamics of a finite-mass mobile electric dipole interacting with various forms of light. In particular, calculations are given for a single monochromatic beam (Sec. III) and counterpropagating beams (Sec. IV). In all cases we show that the new interaction is responsible for changes of conventional results. Furthermore, there are additional effects whose main feature is the scattering of the system in directions transverse to the light beam. Section V contains our main conclusions and we comment further on the possibility of experimentally observing the transverse force,

one of the main manifestations of the Röntgen interaction in the case of irradiation with a single beam.

II. THEORY

A. Canonical formalism

The theory of a mobile dipole is derivable from conventional quantum electrodynamics as follows [6]. We consider an electrically neutral system of two opposite charges e_1 and e_2 of masses m_1 and m_2 in the presence of electromagnetic fields. The conventional nonrelativistic Lagrangian in the radiation gauge is given by

$$L=\tfrac{1}{2}m_1\dot{\mathbf{q}}_1^2+\tfrac{1}{2}m_2\dot{\mathbf{q}}_2^2+\frac{e_1e_2}{4\pi\epsilon_0|\mathbf{q}_1-\mathbf{q}_2|}+\int\mathcal{L}d\mathbf{r}\ , \tag{1}$$

where

$$\mathcal{L}=\mathbf{J}\cdot\mathbf{A}^{\perp}+\tfrac{1}{2}\epsilon_0[\dot{\mathbf{A}}^{\perp 2}-c^2(\nabla\times\mathbf{A}^{\perp})^2] \tag{2}$$

with $\mathbf{q}_1$ and $\mathbf{q}_2$ the particle position vectors and $\dot{\mathbf{q}}_1$ and $\dot{\mathbf{q}}_2$ the corresponding velocities. The Coulomb effects have been separated out, as accounted for by the static interparticle interaction, so there is no scalar potential. The field variable $\mathbf{A}^{\perp}$ is the vector potential in the transverse (radiation) gauge formally defined by the equations

$$\nabla\cdot\mathbf{A}^{\perp}=0\ ,\quad \phi=0\ . \tag{3}$$

In Eq. (2) $\mathbf{J}(\mathbf{r})$ is the total current density

$$\mathbf{J}(\mathbf{r})=e_1\dot{\mathbf{q}}_1\delta(\mathbf{r}-\mathbf{q}_1)+e_2\dot{\mathbf{q}}_2\delta(\mathbf{r}-\mathbf{q}_2)\ . \tag{4}$$

The canonical variables in this framework are $\mathbf{q}_1$, $\mathbf{q}_2$, and $\mathbf{A}(\mathbf{r})$. The corresponding momenta are $\mathbf{p}_1$, $\mathbf{p}_2$, and $\mathbf{\Pi}^{\perp}(\mathbf{r})$,

$$\mathbf{p}_\alpha=\frac{\partial L}{\partial\dot{\mathbf{q}}_\alpha}=m_\alpha\dot{\mathbf{q}}_\alpha-e_\alpha\mathbf{A}^{\perp}(\mathbf{q}_\alpha)\ ,\quad \alpha=1,2\ , \tag{5}$$

$$\mathbf{\Pi}^{\perp}=\frac{\partial\mathcal{L}}{\partial\dot{\mathbf{A}}^{\perp}}=\epsilon_0\dot{\mathbf{A}}^{\perp}=-\epsilon_0\mathbf{E}^{\perp}\ . \tag{6}$$

The commutation relations are

$$[p_{\alpha i},q_{\alpha' j}]=-i\hbar\delta_{\alpha\alpha'}\delta_{ij}\ , \tag{7}$$

$$[\Pi_i^{\perp}(\mathbf{r}),A_j^{\perp}(\mathbf{r}')]=-i\hbar\delta_{ij}^{\perp}(\mathbf{r}-\mathbf{r}')\ , \tag{8}$$

where $\delta_{ij}^{\perp}(\mathbf{r}-\mathbf{r}')$ is the transverse δ function [7]. The corresponding Hamiltonian is

$$H=\dot{\mathbf{q}}_1\cdot\mathbf{p}_1+\dot{\mathbf{q}}_2\cdot\mathbf{p}_2+\int\dot{\mathbf{A}}^{\perp}\cdot\mathbf{\Pi}^{\perp}d\mathbf{r}-L\ . \tag{9}$$

Using $\mathbf{B}=\nabla\times\mathbf{A}^{\perp}$, we obtain

$$H=\frac{[\mathbf{p}_1+e_1\mathbf{A}^{\perp}(\mathbf{q}_1)]^2}{2m_1}+\frac{[\mathbf{p}_2+e_2\mathbf{A}^{\perp}(\mathbf{q}_2)]^2}{2m_2}+\frac{e_1e_2}{4\pi\epsilon_0|\mathbf{q}_1-\mathbf{q}_2|}+\tfrac{1}{2}\int\left[\frac{\mathbf{\Pi}^{\perp 2}}{\epsilon_0}+\frac{\mathbf{B}^2}{\mu_0}\right]d\mathbf{r}\ . \tag{10}$$

It can be checked that the above formalism yields the equations of motion as Maxwell's equations and Newton's law with the Lorentz force. In particular, we have for the latter

$$m_\alpha\ddot{\mathbf{q}}_\alpha=e_\alpha(\mathbf{E}(\mathbf{q}_\alpha)+\dot{\mathbf{q}}_\alpha\times\mathbf{B}(\mathbf{q}_\alpha))\ ,\quad \alpha=1,2\ . \tag{11}$$

We seek to express the Hamiltonian in Eq. (10) in a multipolar form. Our ultimate aim, however, is to arrive at a version of the Hamiltonian which is valid in the electric-dipole approximation. We begin by defining a canonical transformation characterized by the following generating function:

$$S=e^{i\Lambda}=\exp\left[\frac{-i}{\hbar}\int\mathcal{P}(\mathbf{r})\cdot\mathbf{A}^{\perp}(\mathbf{r})d\mathbf{r}\right]\ , \tag{12}$$

where $\mathcal{P}$ is the polarization field relative to the center-of-mass coordinate $\mathbf{R}$,

$$\mathcal{P}(\mathbf{r})=\sum_{\alpha=1,2}e_\alpha(\mathbf{q}_\alpha-\mathbf{R})\int_0^1 d\lambda\delta(\mathbf{r}-\mathbf{R}-\lambda(\mathbf{q}_\alpha-\mathbf{R}))\ , \tag{13}$$

where $\mathbf{R}$ is given by

$$\mathbf{R}=\frac{m_1\mathbf{q}_1+m_2\mathbf{q}_2}{M}\ ,\quad M=m_1+m_2\ . \tag{14}$$

Formally, the effect of the transformation is to give rise to a new Hamiltonian H_{trans} which is identical in appearance to the old Hamiltonian H, except that the momentum variables are primed. We have

$$H_{\text{trans}}=\frac{[\mathbf{p}_1'+e_1\mathbf{A}^{\perp}(\mathbf{q}_1)]^2}{2m_1}+\frac{[\mathbf{p}_2'+e_2\mathbf{A}^{\perp}(\mathbf{q}_2)]^2}{2m_2}+\frac{e_1e_2}{4\pi\epsilon_0|\mathbf{q}_1-\mathbf{q}_2|}+\tfrac{1}{2}\int\left[\frac{\mathbf{\Pi}'^{\perp 2}}{\epsilon_0}+\frac{\mathbf{B}^2}{\mu_0}\right]d\mathbf{r}\ . \tag{15}$$

In this framework we can show that the particle velocities are given by

$$m_\alpha\dot{\mathbf{q}}_\alpha=\mathbf{p}_\alpha'+e_\alpha\mathbf{A}^{\perp}(\mathbf{q}_\alpha)\ . \tag{16}$$

The canonical momenta $\mathbf{p}_\alpha$ and $\mathbf{\Pi}^{\perp}$ are related to the new ones $\mathbf{p}_\alpha'$ and $\mathbf{\Pi}'^{\perp}$ as follows:

$$\mathbf{p}_\alpha'=e^{-i\Lambda}\mathbf{p}_\alpha e^{i\Lambda}=\mathbf{p}_\alpha+i[\mathbf{p}_\alpha,\Lambda]\ , \tag{17}$$

$$\mathbf{\Pi}'^{\perp}=e^{-i\Lambda}\mathbf{\Pi}^{\perp}e^{i\Lambda}=\mathbf{\Pi}^{\perp}+i[\mathbf{\Pi}^{\perp},\Lambda]\ . \tag{18}$$

The series in Eqs. (17) and (18) both terminate at the first commutator on account of the form of Λ in Eq. (12). We have

$$\mathbf{p}_\alpha'=\mathbf{p}_\alpha+\hbar\nabla^\alpha\Lambda\ , \tag{19}$$

$$\mathbf{\Pi}'^{\perp}(\mathbf{r})=\mathbf{\Pi}^{\perp}(\mathbf{r})-\mathcal{P}^{\perp}(\mathbf{r})\ , \tag{20}$$

where ∇^α in Eq. (19) refers to differentiation with respect to the coordinate $\mathbf{q}^\alpha$. In Eq. (18) we have introduced $\mathcal{P}^{\perp}$ as the transverse part of the polarization vector given in Eq. (13), having made use of the commutator in Eq. (8). The gradient $\nabla^\alpha\Lambda$ can be straightforwardly evaluated. We have

$$\hbar\nabla^\alpha\Lambda=-e_\alpha\mathbf{A}^{\perp}(\mathbf{q}_\alpha)-\int\mathbf{\Theta}_\alpha(\mathbf{r})\times(\nabla\times\mathbf{A}^{\perp}(\mathbf{r}))d\mathbf{r}\ . \tag{21}$$

The vector $\mathbf{\Theta}_\alpha$ is given by

$$\Theta_\alpha(\mathbf{r})=\sum_{\beta=1,2} e_\beta \int_0^1 d\lambda(\lambda\delta_{\alpha\beta}-\frac{m_\alpha}{M}[\lambda-1])(\mathbf{q}_\beta-\mathbf{R}) \times\delta(\mathbf{r}-\mathbf{R}-\lambda(\mathbf{q}_\beta-\mathbf{R})) . \quad (22)$$

Note that different versions of $\nabla^\alpha\Lambda$ and Θ_α were quoted in Ref. [5], where the theory was designed to deal with an arbitrary atomic system. In Ref. [5] there was need to introduce equations of constraint in order to preserve the number of degrees of freedom when the center of mass was invoked as an independent dynamical variable. The version of $\nabla^\alpha\Lambda$ given here in Eqs. (21) and (22) is convenient for the two-particle case, the details of which are found elsewhere [8]. The complete multipolar Hamiltonian follows from Eq. (15) by direct use of Eqs. (19) and (20) and the division of the motion into internal plus gross motion can subsequently be carried out. However, since our aim is to obtain the electric-dipole version of the Hamiltonian, it is much more convenient to impose this approximation at this stage.

B. Decoupling of motions in dipole approximation

We introduce the internal variable $\mathbf{q}$ as follows:

$$\mathbf{q}=\mathbf{q}_1-\mathbf{q}_2 , \quad (23)$$

and we also set

$$e_1=-e_2=e . \quad (24)$$

Then clearly we have, using Eq. (14),

$$\mathbf{q}_1-\mathbf{R}=\frac{m_2}{M}\mathbf{q} , \quad \mathbf{q}_2-\mathbf{R}=-\frac{m_1}{M}\mathbf{q} . \quad (25)$$

The electric-dipole approximation corresponds to retaining the leading terms in the expansions of the δ functions appearing in the polarization vectors $\mathcal{P}$ and Θ_α in powers of $q_\alpha-\mathbf{R}$. It is convenient to continue to use the same symbols for the electric-dipole version of various quantities. We have from Eq. (13) after making use of Eqs. (23)–(25),

$$\mathcal{P}=-\mathbf{d}\delta(\mathbf{r}-\mathbf{R}) , \quad (26)$$

where $\mathbf{d}$ is the electric-dipole moment vector of the system

$$\mathbf{d}=-\sum_{\alpha=1,2} e_\alpha(\mathbf{q}_\alpha-\mathbf{R})=-e\mathbf{q} . \quad (27)$$

Similarly, the dipole approximation version of Θ_α is obtained from Eq. (22) after some algebra using Eqs. (23)–(25) in the simple form

$$\Theta_\alpha=-\tfrac{1}{2}\mathbf{d}\delta(\mathbf{r}-\mathbf{R}) , \quad \alpha=1,2 . \quad (28)$$

Substituting in Eq. (21), we have

$$\hbar\nabla^\alpha\Lambda=-e_\alpha\mathbf{A}^\perp(\mathbf{q}_\alpha)+\tfrac{1}{2}\mathbf{d}\times\mathbf{B}(\mathbf{R}) . \quad (29)$$

Hence we can write for the transformed momenta, Eqs. (19) and (20),

$$\mathbf{p}'_\alpha=\mathbf{p}_\alpha-e_\alpha\mathbf{A}^\perp(\mathbf{q}_\alpha)+\tfrac{1}{2}\mathbf{d}\times\mathbf{B}(\mathbf{R}) , \quad (30)$$

$$\mathbf{\Pi}'^\perp(\mathbf{r})=\mathbf{\Pi}^\perp(\mathbf{r})-\mathcal{P}^\perp(\mathbf{r}) . \quad (31)$$

Finally, substituting from Eqs. (30) and (31) in Eq. (15), we obtain the transformed Hamiltonian in the following form:

$$H_{\text{trans}}=\sum_{\alpha=1,2}\frac{[\mathbf{p}_\alpha+\frac{1}{2}\mathbf{d}\times\mathbf{B}(\mathbf{R})]^2}{2m_\alpha}-\frac{e^2}{4\pi\epsilon_0 q} +\frac{1}{2}\int\left\{\frac{[\mathbf{\Pi}^\perp(\mathbf{r})-\mathcal{P}^\perp(\mathbf{r})]^2}{\epsilon_0}+\frac{\mathbf{B}^2(\mathbf{r})}{\mu_0}\right\}d\mathbf{r} . \quad (32)$$

The next steps involve the division of the motion into translational and internal motions. We define the center-of-mass momentum $\mathbf{P}$ conjugate to the center-of-mass position vector $\mathbf{R}$, Eq. (14),

$$\mathbf{P}=\mathbf{p}_1+\mathbf{p}_2 , \quad (33)$$

and the internal momentum $\mathbf{p}$ conjugate to the internal variable $\mathbf{q}$ by

$$\mathbf{p}=\frac{m_2\mathbf{p}_1-m_1\mathbf{p}_2}{M} , \quad \mathbf{q}=\mathbf{q}_1-\mathbf{q}_2 . \quad (34)$$

We can then express $\mathbf{p}_1$ and $\mathbf{p}_2$ in terms of $\mathbf{P}$ and $\mathbf{p}$ as follows:

$$\mathbf{p}_\alpha=\frac{m_\alpha}{M}\mathbf{P}+(-1)^{\alpha+1}\mathbf{p} , \quad \alpha=1,2 . \quad (35)$$

Equation (35) enables the explicit change from the particle canonical variables $\mathbf{q}_\alpha$ and $\mathbf{p}_\alpha$ to internal variables $(\mathbf{q},\mathbf{p})$ and gross motion variables $(\mathbf{R},\mathbf{P})$. That the new pairs are independent canonical variables can easily be checked. We have

$$[P_i,R_j]=-i\hbar\delta_{ij} , \quad (36)$$

$$[p_i,q_j]=-i\hbar\delta_{ij} , \quad (37)$$

$$[P_i,q_j]=0=[p_i,R_j] , \quad (38)$$

which follow by direct use of the commutator

$$[p_{\alpha i},q_{\beta j}]=-i\hbar\delta_{\alpha\beta}\delta_{ij} . \quad (39)$$

The relationships in Eqs. (36), (37), (38), and (39) ensure that the new variables conform with the requirements for independent sets representing two independent motions in the absence of coupling. Substituting from Eq. (35) in Eq. (32), we get

$$H_{\text{trans}}=\sum_{\alpha=1,2}\left[\frac{[(P/M)m_\alpha+(-1)^{\alpha+1}\mathbf{p}+\frac{1}{2}\mathbf{d}\times\mathbf{B}(\mathbf{R})]^2}{2m_\alpha}\right] -\frac{e^2}{4\pi\epsilon_0 q}+\tfrac{1}{2}\int\left[\frac{[\mathbf{\Pi}^\perp(\mathbf{r})-\mathcal{P}^\perp(\mathbf{r})]^2}{\epsilon_0}+\frac{\mathbf{B}^2(\mathbf{r})}{\mu_0}\right]d\mathbf{r} . \quad (40)$$

On expanding the squares we find that the sum in the first term of Eq. (40) simplifies considerably, and we obtain

$$H_{\text{trans}}=\frac{\mathbf{P}^2}{2M}+\left[\frac{\mathbf{p}^2}{2\mu}-\frac{e^2}{4\pi\epsilon_0 q}\right]+\tfrac{1}{2}\int\left[\frac{\mathbf{\Pi}^{\perp 2}(\mathbf{r})}{\epsilon_0}+\frac{B^2(\mathbf{r})}{\mu_0}\right]d\mathbf{r}+\frac{1}{\epsilon_0}\mathbf{d}\cdot\mathbf{\Pi}^{\perp}(\mathbf{R})+\frac{1}{2M}(\mathbf{P}\cdot\mathbf{d}\times\mathbf{B}(\mathbf{R})+\mathbf{d}\times\mathbf{B}(\mathbf{R})\cdot\mathbf{P})$$
$$+\frac{[\mathbf{d}\times\mathbf{B}(\mathbf{R})]^2}{8\mu}+\frac{1}{2\epsilon_0}\int\mathcal{P}^{\perp}(\mathbf{r})^2 d\mathbf{r}+(\text{magnetic-dipole terms}) , \tag{41}$$

where μ is the reduced mass

$$\mu=\frac{m_1 m_2}{M} . \tag{42}$$

The magnetic-dipole terms arise from the product between $\mathbf{p}$ and $\mathbf{d}\times\mathbf{B}$ and, when explicitly written, are seen to lead to the usual form as a purely internal interaction that is negligible in comparison with the electric-dipole interaction [9].

The result given in Eq. (41) is the nonrelativistic Hamiltonian for a system of bound charges in interaction with the transverse radiation field. In this Hamiltonian the internal motion of the charge system is clearly distinguished from the gross motion. It is seen that the zero-order Hamiltonians of the three subsystems (gross motion, internal motion, and fields) are exactly separated off and given by the first three terms of Eq. (41). The rest of the terms represent the interaction in which the three subsystems are coupled. The fourth and fifth terms constitute the leading interaction and will therefore be retained. The sixth term is a diamagnetic-type energy which is relatively small. The term involving the integral of the square of the polarization is a self-energy which is known to contribute to the Lamb shift and may be absorbed in any renormalized energies pertaining to the internal motion.

The effective Hamiltonian H_d is defined as the version of H_{trans} in the electric-dipole approximation and restricted to the following truncated form:

$$H_d=\frac{\mathbf{P}^2}{2M}+H_a+H_f-\mathbf{d}\cdot\mathbf{E}^{\perp}(\mathbf{R})$$
$$+\frac{1}{2M}(\mathbf{P}\cdot\mathbf{d}\times\mathbf{B}(\mathbf{R})+\mathbf{d}\times\mathbf{B}(\mathbf{R})\cdot\mathbf{P}) , \tag{43}$$

where H_a is the hydrogenlike Hamiltonian describing the internal motion while H_f is the field Hamiltonian. In quantized form the field Hamiltonian becomes

$$H_f=\sum_{\lambda}\sum_{\mathbf{k}}\hbar\omega_{k\lambda}(a^{\dagger}_{k\lambda}a_{k\lambda}+\tfrac{1}{2}) , \tag{44}$$

corresponding to quantized $\mathbf{A}^{\perp}$ given by

$$\mathbf{A}^{\perp}(\mathbf{r})=\sum_{\lambda}\sum_{\mathbf{k}}\hat{\epsilon}_{k\lambda}\left[\frac{\hbar}{2\epsilon_0\tau\omega_{k\lambda}}\right]^{1/2}a_{k\lambda}e^{i\mathbf{k}\cdot\mathbf{r}}+\text{H.c.} , \tag{45}$$

where τ is a quantization volume and $\hat{\epsilon}_{k\lambda}$ is a unit polarization vector. Note that in Eq. (43) the electric-dipole interaction is written in the conventional form $-\mathbf{d}\cdot\mathbf{E}^{\perp}(\mathbf{R})$, i.e., in terms of $\mathbf{E}^{\perp}$ rather than $\mathbf{\Pi}^{\perp}$, as it correctly appears in Eq. (41). The importance of distinguishing between the roles of the electric field vector and the true field canonical momentum in various formulations is discussed in detail in Ref. [5]. In the radiation gauge the relationship between the vector fields $\mathbf{E}^{\perp}$ and $\mathbf{\Pi}^{\perp}$ is given by Eq. (6), so that the equal-time commutation relation, Eq. (8), can be written at once as follows:

$$\epsilon_0[E_i^{\perp}(\mathbf{r}),A_j^{\perp}(\mathbf{r}')]=i\hbar\delta_{ij}^{\perp}(\mathbf{r}-\mathbf{r}') . \tag{46}$$

Equation (43) is the starting point for the investigation of gross motion effects. The interesting feature is the appearance of the $\mathbf{P}$-dependent terms, which we refer to as Röntgen-type interaction terms.

C. Mechanical momentum and pressure force

The form of the Hamiltonian in Eq. (41) suggests that an additional vector field which is proportional to $\mathbf{d}\times\mathbf{B}$ plays the role of an electromagnetic vector potential modifying the kinetic-energy term of the Hamiltonian pertaining to the gross motion in a manner that is analogous to the case of a single charge in an $\mathbf{A}$ field. There is therefore a need to distinguish between the canonical and the mechanical momenta of the gross motion.

From Eq. (30) we can write

$$\mathbf{p}'_{\alpha}+e_{\alpha}\mathbf{A}^{\perp}(\mathbf{q}_{\alpha})=\mathbf{p}_{\alpha}+\tfrac{1}{2}\mathbf{d}\times\mathbf{B}(\mathbf{R}) . \tag{47}$$

The left-hand side is precisely $m_{\alpha}\dot{\mathbf{q}}_{\alpha}$ associated with the transformed Hamiltonian and given by Eq. (16). Thus we obtain

$$m_{\alpha}\dot{\mathbf{q}}_{\alpha}=\mathbf{p}_{\alpha}+\tfrac{1}{2}\mathbf{d}\times\mathbf{B}(\mathbf{R}) . \tag{48}$$

Introducing a sum over α in each term, we get

$$\sum_{\alpha=1,2}m_{\alpha}\dot{\mathbf{q}}_{\alpha}=\sum_{\alpha=1,2}\mathbf{p}_{\alpha}+\mathbf{d}\times\mathbf{B}(\mathbf{R}) . \tag{49}$$

On making use of Eq. (14) we find at once that we can identify the left-hand side of Eq. (49) with $M\dot{\mathbf{R}}$ and the first term on the right-hand side with $\mathbf{P}$. Thus we can write

$$M\dot{\mathbf{R}}=\mathbf{P}+\mathbf{d}\times\mathbf{B}(\mathbf{R}) . \tag{50}$$

Equation (50) establishes the difference between the canonical momentum $\mathbf{P}$ and the mechanical momentum $M\dot{\mathbf{R}}$. The result also follows as a Heisenberg operator equation based on the Hamiltonian in Eq. (43). We have, using Eq. (36),

$$\dot{\mathbf{R}}=\frac{i}{\hbar}[H_d,\mathbf{R}]=\frac{\mathbf{P}+\mathbf{d}\times\mathbf{B}(\mathbf{R})}{M} . \tag{51}$$

By analogy with the case of a single charge we expect that the relevant dynamical quantity should be the mechanical momentum $M\dot{\mathbf{R}}$ rather than the canonical one $\mathbf{P}$. Once this important distinction is established the radiation pressure force on the dipole follows from Eq.

(50) as follows:

$$\mathbf{F}=\frac{d}{dt}(M\dot{\mathbf{R}})=\frac{d\mathbf{P}}{dt}+\frac{d}{dt}(\mathbf{d}\times\mathbf{B}) . \tag{52}$$

The force also follows from Eq. (51) as a Heisenberg operator equation in the form

$$M\ddot{\mathbf{R}}=\frac{i}{\hbar}[H_d,(\mathbf{P}+\mathbf{d}\times\mathbf{B})]$$

$$=\nabla(\mathbf{d}\cdot\mathbf{E}(\mathbf{R}))+\frac{d}{dt}(\mathbf{d}\times\mathbf{B}) , \tag{53}$$

where ∇ refers to differentiation with respect to the components of $\mathbf{R}$ and we have explicitly evaluated the first commutator $(i/\hbar)[H_d,\mathbf{P}]=\nabla(\mathbf{d}\cdot\mathbf{E})$ but left the second commutator as a time derivative.

The result in Eq. (53) is consistent with a simple derivation [10] based on the Lorentz force expression [Eq. (11)],

$$\mathbf{F}=\sum_{\alpha=1,2} e_\alpha(\mathbf{E}(\mathbf{q}_\alpha)+\dot{\mathbf{q}}_\alpha\times\mathbf{B}(\mathbf{q}_\alpha)) . \tag{54}$$

In the dipole approximation and making use of Eqs. (23), (24), and (27) we have by Taylor expansion

$$\sum_{\alpha=1,2} e_\alpha\mathbf{E}(\mathbf{q}_\alpha)\approx(\mathbf{d}\cdot\nabla)\mathbf{E}$$

$$=\nabla(\mathbf{d}\cdot\mathbf{E})-\mathbf{d}\times\nabla\times\mathbf{E}$$

$$=\nabla(\mathbf{d}\cdot\mathbf{E})+\mathbf{d}\times\frac{\partial\mathbf{B}}{\partial t} . \tag{55}$$

Similarly, we can write

$$\sum_\alpha e_\alpha\dot{\mathbf{q}}_\alpha\times\mathbf{B}(\mathbf{q}_\alpha)\approx\frac{\partial\mathbf{d}}{\partial t}\times\mathbf{B} . \tag{56}$$

Thus we obtain using Eqs. (55) and (56),

$$\mathbf{F}\approx\nabla(\mathbf{d}\cdot\mathbf{E})+\frac{d}{dt}(\mathbf{d}\times\mathbf{B}) . \tag{57}$$

We have therefore established that both dynamical attributes, namely the gross momentum and the corresponding pressure force, receive contributions which are directly attributable to the inclusion of the Röntgen term. The complete Hamiltonian in the dipole approximation (including the Röntgen term) is essential for studying the time evolution of the system. In the next sections we carry out calculations that explore the implications of the formalism developed so far for the dynamics of specific physical situations involving interaction of various forms of light with mobile systems of charges.

III. ATOMIC SYSTEMS IN SINGLE BEAMS

A. Derivation of $\langle M\dot{\mathbf{R}}\rangle$ and $\langle\mathbf{F}\rangle$

We consider a mobile atom with its internal motion subject to the two-level approximation involving only a ground state $|g\rangle$ and an excited state $|e\rangle$. Both states are eigenstates of H_a and are separated by an energy $\hbar\omega_0$. The system is taken to be in interaction with a single laser mode of the transverse radiation field of wave vector $\mathbf{k}$, polarization $\hat{\boldsymbol{\epsilon}}$, and frequency ω_k. With this mode we associate the usual annihilation and creation operators a_k and $a_k^\dagger$. The atomic operators are defined by [11]

$$\pi^\dagger=|e\rangle\langle g| , \quad \pi=|g\rangle\langle e| \tag{58}$$

such that the anticommutator holds

$$\{\pi^\dagger,\pi\}=1 . \tag{59}$$

The Hamiltonian of this system corresponding to Eq. (43) can now be written as a Heisenberg operator

$$H_d=\frac{P^2}{2M}+\hbar\omega_0\pi^\dagger\pi+\hbar\omega_k a_k^\dagger a_k+H_{\rm I}+H_{\rm II} , \tag{60}$$

where

$$H_{\rm I}(\mathbf{R},t)=-\mathbf{d}\cdot\mathbf{E}^\perp(\mathbf{R},t) , \tag{61}$$

$$H_{\rm II}(\mathbf{R},t)=\frac{1}{2M\omega_k}\{(\mathbf{d}\times[\mathbf{k}\times\mathbf{E}^\perp])\cdot\mathbf{P}$$

$$+\mathbf{P}\cdot(\mathbf{d}\times[\mathbf{k}\times\mathbf{E}^\perp])\} \tag{62}$$

with $\mathbf{E}^\perp$ given by

$$\mathbf{E}^\perp(\mathbf{R},t)=-\dot{\mathbf{A}}^\perp(\mathbf{R},t)$$

$$=i\left[\frac{\hbar\omega_k}{2\epsilon_0\tau}\right]^{1/2}\hat{\boldsymbol{\epsilon}}[a_k(t)e^{i\mathbf{k}\cdot\mathbf{R}(t)}-\text{H.c.}] . \tag{63}$$

We are interested in the evaluation of the expectation value of the mechanical momentum $\langle M\dot{\mathbf{R}}\rangle$ and of the force $\langle M\ddot{\mathbf{R}}\rangle$ as functions of time and we restrict treatment further to the rotating wave approximation. Consider first the expectation value of the momentum defined by

$$\langle M\dot{\mathbf{R}}(t)\rangle=\frac{iM}{\hbar}\langle\psi|e^{iH_d t/\hbar}[H_d,\mathbf{R}(t)]e^{-iH_d t/\hbar}|\psi\rangle , \tag{64}$$

where ψ is a general state of the overall system in the absence of the coupling. The commutator in Eq. (64) is obtained straightforwardly as follows:

$$[H_d,\mathbf{R}(t)]=\frac{-i\hbar}{M}\left\{\mathbf{P}+\frac{\mathbf{d}\times[\mathbf{k}\times\mathbf{E}^\perp]}{\omega_k}\right\} . \tag{65}$$

Thus we have from Eq. (64)

$$\langle M\dot{\mathbf{R}}\rangle=\left\langle\psi\left|e^{iH_d t/\hbar}\left\{\mathbf{P}+\frac{\mathbf{d}\times[\mathbf{k}\times\mathbf{E}^\perp]}{\omega_k}\right\}e^{-iH_d t/\hbar}\right|\psi\right\rangle . \tag{66}$$

The next steps require use of the standard expansion of the exponential operators and collecting terms. To leading order and ignoring all counter rotating terms, we have

$$\langle M\dot{\mathbf{R}}(t)\rangle=M\dot{\mathbf{R}}(0)$$

$$+\left\langle\psi\left|\frac{i}{\hbar}\int_0^t\left[H_d,\left[\mathbf{P}+\frac{\mathbf{d}\times[\mathbf{k}\times\mathbf{E}^\perp]}{\omega_k}\right]\right]dt\right|\psi\right\rangle . \tag{67}$$

The procedure requires first calculations of the commuta-

tor in Eq (67) using Eq. (60), keeping terms up to order $1/M$, then substituting the appropriate time dependence of the operators $\mathbf{R}(t)$, $a(t)$, $\pi(t)$, etc. We obtain after much algebra the following result:

$$\langle M\dot{\mathbf{R}}(t)\rangle = M\mathbf{V}+\hbar k\{G_k\hat{\mathbf{k}}+U_k\hat{\boldsymbol{\epsilon}}\} \times[-n_e-n_k(2n_e-1)]I(t)\ , \quad (68)$$

where $\hat{\mathbf{k}}$ is a unit vector in the direction of $\mathbf{k}$ and $\mathbf{V}$ is the initial velocity

$$\mathbf{V}=\dot{\mathbf{R}}(0)\ . \quad (69)$$

The functions G_k and U_k are given by

$$G_k=\frac{\omega_0}{\omega_k}g_k(g_k+S_k(\mathbf{V}))\ , \quad (70)$$

$$U_k=\left[1-\frac{\omega_0}{\omega_k}\right]f_k(g_k+T_k(\mathbf{V})) \quad (71)$$

with g_k and f_k the velocity-independent factors

$$g_k=(\mathbf{d}\cdot\hat{\boldsymbol{\epsilon}})\left[\frac{\omega_k}{\hbar\epsilon_0\tau}\right]^{1/2}, \quad (72)$$

$$f_k=(\mathbf{d}\cdot\hat{\mathbf{k}})\left[\frac{\omega_k}{\hbar\epsilon_0\tau}\right]^{1/2}, \quad (73)$$

while S_k and T_k depend on the velocity and are given by

$$S_k=-g_k\frac{\mathbf{k}\cdot\mathbf{V}}{\omega_k}+kf_k\hat{\boldsymbol{\epsilon}}\cdot\mathbf{V}\left[\frac{1}{\omega_k}+\frac{1}{\omega_0}\right], \quad (74)$$

$$T_k=-g_k\frac{\mathbf{k}\cdot\mathbf{V}}{\omega_k}+kf_k\frac{\hat{\boldsymbol{\epsilon}}\cdot\mathbf{V}}{\omega_k}-g_k\frac{\mathbf{k}\cdot\mathbf{V}}{\omega_{\mathbf{k}}-\omega_0}\ . \quad (75)$$

The time dependence in Eq. (68) is entirely contained in the function $I(t)$ which is defined by

$$I(t)=\int_0^t\frac{\sin\Delta t}{\Delta}dt =\frac{1}{\Delta^2}\{1-\cos\Delta t\}\ , \quad (76)$$

where Δ is given by

$$\Delta=\omega_0-\omega_k-\mathbf{k}\cdot\mathbf{V}\ . \quad (77)$$

We have also assumed that

$$\langle\psi|\pi^\dagger(0)|\psi\rangle=0=\langle\psi|\pi(0)|\psi\rangle\ . \quad (78)$$

The symbols n_g and n_e are the initial occupation probabilities for the ground state and the excited state, respectively,

$$n_g=\langle\psi|\pi(0)\pi^\dagger(0)|\psi\rangle\ ,\quad n_e=\langle\psi|\pi^\dagger(0)\pi(0)|\psi\rangle\ , \quad (79)$$

such that

$$n_e+n_g=1\ . \quad (80)$$

Finally, n_k is the mean photon number

$$n_k=\langle\psi|a_k^\dagger(0)a_k(0)|\psi\rangle\ . \quad (81)$$

Equation (68) is one of the main results of this paper. It gives the mechanical momentum vector that an atom, of excitation frequency ω_0, will acquire at time t as a result of interaction with the laser mode of frequency ω_k, wave vector $\mathbf{k}$ and polarization $\hat{\boldsymbol{\epsilon}}$.

Next, consider the derivation of the corresponding pressure force. This is defined by

$$\langle\mathbf{F}_{\hat{\mathbf{k}},\hat{\boldsymbol{\epsilon}}}\rangle=\langle M\ddot{\mathbf{R}}\rangle=\left\langle\frac{d}{dt}M\dot{\mathbf{R}}\right\rangle\ . \quad (82)$$

The analog of Eq. (64) is

$$\langle M\ddot{\mathbf{R}}\rangle=-\frac{M}{\hbar^2}\langle\psi|e^{iH_dt/\hbar}[H_d,[H_d,\mathbf{R}(t)]]e^{-iH_dt/\hbar}|\psi\rangle\ . \quad (83)$$

We may therefore follow an analogous procedure to the one that led to Eq. (68). We obtain

$$\langle\mathbf{F}_{\hat{\mathbf{k}},\hat{\boldsymbol{\epsilon}}}\rangle=\hbar k\{G_k\hat{\mathbf{k}}+U_k\hat{\boldsymbol{\epsilon}}\}[-n_e-n_k(2n_e-1)]\frac{\sin\Delta t}{\Delta}\ . \quad (84)$$

Direct comparison with Eq. (68) reveals that the following simple relationship holds:

$$\langle\mathbf{F}_{\hat{\mathbf{k}},\hat{\boldsymbol{\epsilon}}}\rangle=\frac{\partial}{\partial t}\langle M\dot{\mathbf{R}}(t)\rangle\ . \quad (85)$$

Equation (84) generalizes previous results of the pressure force and is the second main result of this paper.

The results can now be discussed for a number of special cases involving irradiation with single and counterpropagating beams.

B. Velocity-independent terms

1. *The large-t limit*

Consider first the case in which we ignore the velocity-dependent terms. This corresponds to setting $\mathbf{V}$, S_k, and T_k to zero in Eqs. (68), (74), (75), and (77). We then see from Eq. (68) that there are two components to the atomic momentum acquired by the interaction with radiation, instead of the single conventional component in the direction of $\mathbf{k}$. There are in fact two new contributions, one modifies the conventional result in the direction of $\mathbf{k}$ and the second is in a direction transverse to $\mathbf{k}$. Both modifications are direct consequences of the inclusion of the Röntgen-type interaction term in the calculations.

The time dependence of $\langle M\dot{\mathbf{R}}\rangle$ is determined by $I(t)$, Eq. (76). The oscillations at large t average to zero leaving only the time-independent part as the gross momentum change due to interaction with light. It can be checked on assuming that $n_e=0$ that the t-independent part is identical to that obtained using energy arguments [12].

Consider next the force as given by Eq (84) on ignoring the velocity-dependent terms. We assume that in the large-t limit we can write

$$\lim_{t\to\infty}\frac{\sin\Delta_0 t}{\Delta_0}\to\frac{\Gamma}{\Gamma^2+\Delta_0^2}\ , \quad (86)$$

where

$$\Delta_0=(\omega_0-\omega_k) \tag{87}$$

and Γ is the linewidth of the atomic transition. Thus we have for the velocity-independent pressure force on the atom

$$\langle \mathbf{F}_{\hat{\mathbf{k}},\hat{\epsilon}} \rangle = n_k \Gamma \hbar k \left\{ \frac{G_k \hat{\mathbf{k}} + U_k \hat{\epsilon}}{\Gamma^2+\Delta_0^2} \right\}, \quad t \to \infty \ . \tag{88}$$

The result in Eq. (88) can now be compared with conventional results [13]. First, we note that, just as in the case of the momentum, there are two components to the pressure force acquired by interaction with radiation. There are two new contributions which are directly attributable to the inclusion of the Röntgen terms, one modifies the old result in the direction of $\mathbf{k}$ and the second is in a direction that is transverse to $\mathbf{k}$. It is easy to check that at exact resonance the new contributions vanish.

From the above formalism we can proceed to derive rate-equation results applicable in the case of broadband incident light in which n_k varies slowly with ω_k across a distribution of transition frequencies. A discussion of this special case beginning with the less general result analogous to Eq. (88) is given by Al-Hilfy and Loudon [14].

2. *Transverse off-resonance contributions*

The above discussion has established that in off-resonance conditions and with velocity-dependent terms ignored, there are contributions to both the momentum and the pressure force that are in the direction of $\hat{\epsilon}$. At first sight this looks improbable since one expects that any component in the direction of the polarization should average to zero. However, it is seen with the use of Eqs. (71), (72), (73), and (88) that this part of the force $\mathbf{F}$ is proportional to

$$(\mathbf{d}\cdot\mathbf{k})(\mathbf{d}\cdot\hat{\epsilon})\hat{\epsilon} \ .$$

Thus $\hat{\epsilon}$ occurs twice, and any tendency to average to zero can be overcome for suitable vector orientations. This contribution will be referred to as the *transverse* force to distinguish it from the *longitudinal* contribution represented by the term in the direction of $\mathbf{k}$ in the force $\mathbf{F}$ given by (88).

Consider the example of the light beam that propagates along the z axis with linear polarization parallel to the x axis. In this case we have

$$(\mathbf{d}\cdot\mathbf{k})(\mathbf{d}\cdot\hat{\epsilon})\hat{\epsilon}=(d_x d_z,0,0) \ , \tag{89}$$

and there is a nonvanishing transverse force whenever the transition dipole moment $\mathbf{d}$ has nonzero components in both the x and the z directions. The sign of the force clearly depends on the signs of these components. There are, however, quite stringent requirements on the electronic initial and final states that must be satisfied if the transverse force is not to vanish. It is, for example, easy to see that the force is zero for an isotropic atom, where the dipole moment, e.g., for an $s \to p$ transition, can always be chosen parallel to the polarization ϵ of the radiation field.

The transverse force does not vanish, however, for electronic states that have an imposed spatial anisotropy, as, for example, in a beam of appropriately oriented molecules. The required anisotropy can also be achieved, at least in principle, for a double resonance experiment with an atomic beam in which atoms are first excited to a p state and selectively filtered so as to remove all except one of the degenerate orientations; if the filtered atomic beam now encounters an appropriately polarized light beam, it experiences a nonzero transverse force.

It is seen from Eqs. (71) and (88) that the transverse force is directly proportional to the detuning, so that both the magnitude and sign depend on the magnitude and sign of Δ_0. However, the force $\mathbf{F}$ in Eq. (88) is also inversely proportional to $\Gamma^2+\Delta_0^2$, so that the transverse force takes its maximum value for a detuning of order Γ. By contrast, the longitudinal force takes its maximum value for zero detuning, when it is larger than the maximum value of the transverse force by a factor of ω_0/Γ. The transverse force thus has a relatively modest size, in addition to the rather carefully designed experiment that is needed to be able to observe it in principle. Nevertheless, the existence of the transverse force is clearly established by our calculations, and its experimental verification will no doubt occur as measurement techniques continue to develop.

C. Velocity-dependent contributions

Besides the initial mechanical momentum $M\mathbf{V}$ appearing in Eq. (68) the dependence of the results on the initial velocity $\mathbf{V}$ is contained in the functions S_k and T_k as given explicitly by Eqs. (74) and (75). There are further velocity terms entering through the dependence in Eq. (68) on Δ as contained in $I(t)$ and explicitly in Eq. (84). As expected, the magnitudes of the velocity-dependent effects are largely determined by the angular orientation of the particle motion relative to the beam propagation and light polarization. This clearly means that in an ensemble of such a system, atoms moving in different directions relative to a fixed beam configuration sample different velocity-dependent contributions. However, for a fixed atomic beam the presence of the velocity-dependent terms can, in principle, lead to additional effects whose importance increases with velocity. They are prominent in the particular arrangements in which the velocity-independent terms are effectively cancelled out as we discuss next for the case of counterpropagating light beams.

IV. COUNTERPROPAGATING BEAMS

In principle, the atomic system can be subject to irradiation with an arbitrary configuration of laser beams. We continue to exclude the high-field regime and assume that the system is amenable to analysis in terms of perturbation theory as applicable to the single-beam case. Then the effects of irradiation by a complex configuration of light beams can be analyzed using the superposition principle. The case involving two beams of light propagating

in opposite directions is one of the most widely discussed configurations in the field of gross-motion dynamics. An atom situated between the sources experiences effects arising from both as simply the sum of their individual effects. This configuration is the basis of the effect known as one-dimensional optical molasses leading to laser cooling [15].

Consider for convenience only the force arising from interaction of the atomic system discussed here with two such linearly polarized counterpropagating beams. The beams normally have the same frequency ω_k, but their directions of propagation are taken as $\hat{\mathbf{k}}$ and $-\hat{\mathbf{k}}$, and the corresponding linear polarization vectors may either be the same for both, i.e., $\hat{\epsilon}$, or in opposite directions, i.e., $\hat{\epsilon}$ and $-\hat{\epsilon}$. The general case of interest may involve beams having different frequencies and general polarization vectors. We can then write the average force as the following sum:

$$\langle \mathbf{F}_{\text{total}} \rangle = \langle \mathbf{F}_{\hat{\mathbf{k}},\hat{\epsilon}} \rangle + \langle \mathbf{F}_{-\hat{\mathbf{k}},\hat{\epsilon}'} \rangle , \tag{90}$$

where the first term arises from interaction with the beam of frequency ω_k and polarization $\hat{\epsilon}$ propagating in the direction $\hat{\mathbf{k}}$, and the second from that with polarization vector $\hat{\epsilon}'$ and frequency $\omega_{k'}$ propagating in the direction of $-\hat{\mathbf{k}}$.

Substituting from Eq. (84), we obtain

$$\langle \mathbf{F}_{\text{total}} \rangle = \hbar k \left\{ [G_k \hat{\mathbf{k}} + U_k \hat{\epsilon}] \frac{\sin \Delta t}{\Delta} + [-G_{-k'} \hat{\mathbf{k}} + U_{-k'} \hat{\epsilon}'] \frac{\sin \Delta' t}{\Delta'} \right\} \times (-n_e - n_k [2n_e - 1]) , \tag{91}$$

where for convenience we set $n_{k'} = n_k$. The functions $\Delta', G_{-k'}$, and $U_{-k'}$ are defined by

$$\Delta' = \omega_0 - \omega_{k'} + \mathbf{k}' \cdot \mathbf{V} = \Delta_0' + \mathbf{k}' \cdot \mathbf{V} , \tag{92}$$

$$G_{-k'} = \frac{\omega_0}{\omega_{k'}} g_{k'} (g_{k'} - S_{k'}(\mathbf{V})) , \tag{93}$$

$$U_{-k'} = -\left[1 - \frac{\omega_0}{\omega_{k'}} \right] f_{k'} (g_{k'} - T_{k'}(\mathbf{V})) , \tag{94}$$

where we have made use of the identities

$$g_{-k'} = +g_{k'} , \quad f_{-k'} = -f_{k'} , \quad S_{-k'} = -S_{k'} , \quad T_{-k'} = -T_{k'} . \tag{95}$$

In the large-t limit the time dependence in Eq. (91) can be handled in an analogous manner to that in Eq. (88). We obtain, from Eq. (91), in the large-t limit, a result which we can rearrange as follows:

$$\langle \mathbf{F}_{\text{total}} \rangle = \{ -n_e - n_k [2n_e - 1] \} (\mathbf{F}_{\mathbf{k}} + \mathbf{F}_{\epsilon}) , \tag{96}$$

where

$$\mathbf{F}_{\mathbf{k}} = \hbar k \Gamma \left[\frac{G_k}{\Gamma^2 + (\Delta_0 - \mathbf{k} \cdot \mathbf{V})^2} - \frac{G_{-k'}}{\Gamma^2 + (\Delta_0' + \mathbf{k}' \cdot \mathbf{V})^2} \right] \hat{\mathbf{k}} , \tag{97}$$

$$\mathbf{F}_{\epsilon} = \hbar k \Gamma \left[\frac{U_k \hat{\epsilon}}{\Gamma^2 + (\Delta_0 - \mathbf{k} \cdot \mathbf{V})^2} + \frac{U_{-k'} \hat{\epsilon}'}{\Gamma^2 + (\Delta_0' + \mathbf{k}' \cdot \mathbf{V})^2} \right] . \tag{98}$$

Equation (96) has a number of interesting special cases which we consider next.

A. Optical molasses

The simplest case is when the beams have the same frequency $\omega_k = \omega_{k'}$, (i.e., $\Delta_0 = \Delta_0'$) and the same polarizations $\hat{\epsilon} = \hat{\epsilon}'$. This is the configuration normally considered, leading to optical molasses.

Furthermore, we consider the limit in which either the velocity is small in magnitude or its orientation is such that $\mathbf{k} \cdot \mathbf{V} \ll \Delta_0$. This limit corresponds to the following expressions for $\mathbf{F}_{\mathbf{k}}$ and $\mathbf{F}_{\epsilon}$:

$$\mathbf{F}_{\mathbf{k}} = 2\hbar k \Gamma \frac{(\omega_0/\omega_k)}{\Gamma^2 + \Delta_0^2} \left[\frac{2(\mathbf{k} \cdot \mathbf{V}) g_k^2 \Delta_0}{\Gamma^2 + \Delta_0^2} - S_k g_k \right] \hat{\mathbf{k}} \tag{99}$$

and

$$\mathbf{F}_{\epsilon} = -\hbar k \Gamma \frac{(2\Delta_0/\omega_k)}{\Gamma^2 + \Delta_0^2} \left[\frac{2(\mathbf{k} \cdot \mathbf{V}) \Delta_0 g_k f_k}{\Gamma^2 + \Delta_0^2} + T_k f_k \right] \hat{\epsilon} , \tag{100}$$

where we have retained only terms up to those that are linear in the velocity in the expansion of the denominators in powers of $\mathbf{k} \cdot \mathbf{V} / \Delta_0$.

It can now be easily seen that the first term in Eq. (99) corresponds to the well-known friction force in one-dimensional optical molasses [15]. Clearly, with $\mathbf{V}$ assumed to be in an arbitrary direction we have here a theory that yields a generalized friction force. Moreover, we have additional terms that account for the velocity orientational dependence of the force in the direction of propagation $\hat{\mathbf{k}}$. For velocities almost perpendicular to the propagation direction, the friction force is dominated by the last term in Eq. (99). There are also new terms arising in the direction of $\hat{\epsilon}$ which completely dominate the total force when the dipole is parallel to the propagation direction. We comment further on these results in the final section.

The opposite regime $\Delta_0 \ll \mathbf{k} \cdot \mathbf{V}$ in the optical molasses configuration considered above corresponds to the following expressions for $\mathbf{F}_{\mathbf{k}}$ and $\mathbf{F}_{\epsilon}$:

$$\mathbf{F}_k = 2\hbar k \Gamma \frac{(1 + \Delta_0/\omega_k)}{\Gamma^2 + (\mathbf{k} \cdot \mathbf{V})^2} \left[\frac{2(\mathbf{k} \cdot \mathbf{V}) g_k^2 \Delta_0}{\Gamma^2 + (\mathbf{k} \cdot \mathbf{V})^2} - S_k g_k \right] \hat{\mathbf{k}} \tag{101}$$

and

$$\mathbf{F}_\epsilon = -\hbar k \Gamma \frac{(2\Delta_0/\omega_k)}{\Gamma^2+(\mathbf{k}\cdot\mathbf{V})^2}\left[\frac{2(\mathbf{k}\cdot\mathbf{V})\Delta_0 g_k f_k}{\Gamma^2+(\mathbf{k}\cdot\mathbf{V})^2}+T_k f_k\right]\hat{\boldsymbol{\epsilon}}\ . \tag{102}$$

The above expressions are applicable at high velocities, corresponding, for example, to the early stages of laser cooling.

B. Optical molasses for opposite polarizations

It is easy to check that on reversing the signs of both $\hat{\boldsymbol{\epsilon}}$ and $\mathbf{k}$ both f_k and g_k change sign. The corresponding functions G_{-k} and U_{-k}, however, will have the same form as G_k and U_k. In this case we obtain the analogs of Eqs. (99) and (100),

$$\mathbf{F}_k = 4\hbar k \Gamma \frac{(1+\Delta_0/\omega_k)(\mathbf{k}\cdot\mathbf{V})g_k^2\Delta_0}{(\Gamma^2+\Delta_0^2)^2}\hat{\mathbf{k}} \tag{103}$$

and

$$\mathbf{F}_\epsilon = -\hbar k \Gamma \frac{4\Delta_0^2(\mathbf{k}\cdot\mathbf{V})g_k f_k}{\omega_k(\Gamma^2+\Delta_0^2)^2}\boldsymbol{\epsilon}\ . \tag{104}$$

The above result, Eq. (103), shows that it is this configuration that gives rise to the usual friction force at low velocities. In addition, there is a new friction force, Eq. (104), in the transverse direction.

V. COMMENTS AND CONCLUSIONS

The primary concern in this paper has been the gross-motion effects arising in the near-resonance interaction of a two-level system with electromagnetic fields. A simple formalism was developed in Sec. II for a neutral two-particle system as a prototype atomic dipole in the presence of electromagnetic fields. This formalism was shown to lead transparently to the appropriate quantum electrodynamic description of the system, especially for the purpose of investigating the gross-motion effects. The focus of the results is on the Röntgen-type additional terms as shown in Eq. (43). The subsequent calculations are of interest, especially as we have shown in a previous paper [5], because the Röntgen-type effects are important for correctly accounting for the energy-momentum properties of the whole system. It then seems natural to investigate their dynamical consequences in possible experimental setups.

The first task has involved evaluations of the expectation values of the mechanical momentum and of the pressure force appropriate for the two-level atom interacting with a single monochromatic beam of light. The results as given by Eqs. (68) and (84) are general as far as the various parameters of the system are concerned; they are applicable to the case of arbitrary velocity directions and arbitrary dipole orientations. They are therefore in a convenient form for further applications, for example, for ensemble averages involving a collection of such atoms moving in all possible directions and whose dipoles have all possible orientations.

One of the conclusions arising from our results is that the momentum changes and pressure forces due to the irradiation are not confined to the direction of light beam propagation. Components in transverse directions exist, in general, which have velocity-dependent as well as velocity-independent parts, as can be seen by inspecting the expression U_k of Eq. (71). The velocity-independent parts are directly proportional to the detuning Δ_0 defined as the difference between the atomic excitation frequency ω_0 and the frequency of light ω_k. Such velocity-independent terms vanish, therefore, at exact resonance for which $\Delta_0=0$. Nevertheless, the presence of such terms away from resonance is a prediction of the theory which stems directly from the inclusion of the Röntgen-type interaction terms in the formalism and which in turn arise from the requirement of energy-momentum invariance of the whole (atom-radiation) system. The velocity-dependent terms arising in the single-light-beam case are smaller in magnitude than the velocity-independent ones. Note that a residual transverse velocity-dependent contribution exists even at exact resonance. However, in the context of an experiment of the type envisaged here (see Sec. III B 1), the velocity-dependent terms are expected to be negligible, except, perhaps, at high velocities.

By contrast, velocity-dependent effects provide the principal mechanism for modifying atomic gross motion in the case of counterpropagating beams, which constitute the second main application of the formalism in this paper. We have argued that in the perturbative regime, the results for an arrangement involving two counterpropagating beams acting simultaneously on a two-level atom are obtainable simply by adding effects from individual beams. The general results are given by Eqs. (96)–(98). For two identical counterpropagating beams the velocity-independent effects arising from the single-beam arrangement are effectively made to cancel out, leaving only velocity-dependent terms. Of these, the leading terms are attributable to the Doppler effect and the corresponding force, conventionally known as the friction force at low velocities, is directed along the common axis defined by the propagation vector of one of the beams. The results for this case too are obtained as generalized results in the sense that they are applicable to arbitrary velocities and dipole orientations. There are also additional terms arising from the corresponding transverse effects in the single-beam arrangement and whose influence is important when the velocities are high.

Finally, we focus attention on the main consequence of the Röntgen interaction which is manifest in the single-beam case, namely the existence of a transverse off-resonance contribution to the pressure force. We also comment on its possible experimental observation. We have seen that this transverse force, in principle, exists for electronic states that exhibit spatial anisotropy, as for instance in a beam of appropriately oriented molecules [16]. The required anisotropy can also be achieved, at least in principle, for a double resonance experiment involving, for example, $s \rightarrow p$ transition in Rydberg atoms

[17], which are first excited to a p state and then selectively filtered so as to remove all except one of the degenerate orientations. If the filtered atomic beam now encounters an appropriately polarized beam of light, it experiences a nonzero transverse force. It is of considerable interest to see if such a picture can be realized in future experiments.

Note added in proof. Recent work by Wilkens revealed the presence of spurious velocity-dependent terms in the calculated spontaneous emission rate of moving atoms if only the conventional $-\mathbf{d}\cdot\mathbf{E}^{\perp}(\mathbf{R})$ is used [18]. Wilkens subsequently proved (in agreement with our work) that such nonphysical effects vanish only if the Röntgen interaction is included [19]. We would like to thank Dr. Wilkens for drawing our attention to this point.

ACKNOWLEDGMENTS

We would like to thank Professor L. Allen and Dr. A.J. Bain for useful discussions. Thanks are also due to the United Kingdom Science and Engineering Research Council for the financial support of C.B. and V.E.L.

[1] V. G. Minogin and V. S. Letokhov, *Laser Light Pressure on Atoms* (Gordon and Breach, New York, 1987).
[2] A. Ashkin, Phys. Rev. Lett. **24**, 156 (1970); **25**, 1321 (1970).
[3] J. Dalibard and C. Cohen-Tannoudji, J. Opt. Soc. Am. **6**, 2023 (1989).
[4] T. Walker, D. Sesko, and C. E. Wieman, Phys. Rev. Lett. **64**, 408 (1990).
[5] C. Baxter, M. Babiker, and R. Loudon, Phys. Rev. A **47**, 1278 (1993).
[6] C. Baxter, M. Babiker, and R. Loudon, J. Mod. Opt. **37**, 685 (1990).
[7] E. A. Power, *Introductory Quantum Electrodynamics* (Longmans, London, 1964).
[8] V. E. Lembessis (unpublished).
[9] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Springer-Verlag, Berlin, 1957).
[10] S. Stenholm, Rev. Mod. Phys. **58**, 699 (1986).
[11] R. Loudon, *The Quantum Theory of Light*, 2nd ed. (Clarendon, Oxford, 1983).
[12] M. Babiker, J. Phys. B **17**, 4877 (1984). Note, however, that there is a printing error in Eq. (24) of this paper. The correct equation should have a factor of 2 multiplying the second (ϵ) term on the right-hand side of this equation.
[13] J. L. Piqué and J. L. Vialle, Opt. Commun. **5**, 402 (1972).
[14] A. Al-Hilfy and R. Loudon, Opt. Acta **32**, 995 (1985).
[15] W. D. Phillips, in *Fundamental Systems in Quantum Optics*, Proceedings of the International School of Physics, Les Houches, Session LIII, 1990, edited by J. Dalibard, J.-M. Raimond, and J. Zinn-Justin (Elsevier, Amsterdam, 1992), p. 165.
[16] J. R. Rost, J. C. Griffin, B. Friedrich, and D. R. Herschbach, Phys. Rev. Lett. **68**, 1299 (1992).
[17] E. A. Hinds, in *Advances in Atomic and Molecular Physics*, edited by D. Bates and B. Bederson (Academic, New York, 1985), Vol. 20, p. 347.
[18] M. Wilkens, Phys. Rev. A **47**, 671 (1993).
[19] M. Wilkens (unpublished).