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Canonical approach to photon pressure

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A theory of radiation pressure and its effects on the gross motion of an aggregrate of charges is established within a canonical framework. The theory indicates the existence of radiation-pressure effects arising from an additional interaction—the Röntgen interaction—whose origin is the classical Röntgen current. A careful development of the formalism reveals the importance of distinguishing between the canonical and mechanical momenta of the gross motion of the aggregate and of incorporating the Röntgen-type interaction terms when calculating the gross-motion dynamics. A form of the Röntgen interaction is present even in the dipole approximation: an effect which has previously been ignored but which is necessary to ensure gauge invariance of the radiation-induced mechanical force. Explicit calculations of the rates of change of canonical and mechanical momenta are presented for a general atomic dipole, with specializations to a two-level atom, revealing the presence of velocity-dependent terms in a natural way. The formalism is consistent with the Minkowski form of the classical momentum density of an electromagnetic field.

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I. INTRODUCTION

The present strong interest in the mechanical effects of radiation dates from the first observation [1] of the acceleration of microsized particles by laser light in 1970, prompting the suggestion [2] later in the same year that similar effects might be observed at the atomic level using laser light tuned to a particular optical transition. The subsequent fast pace of the experimental work undertaken in the areas of laser cooling and trapping is reflected in the frequent reviews [3].

The object of the present paper is to construct a canonical formalism in which to describe the mechanical effects of radiation, laying the foundation for a rigorous description of the changes in atomic gross motion caused by an interaction with a quantized radiation field. A specific feature of the canonical formalism, which, in the past, has either been ignored or overlooked, is shown to be important in the determinations of radiation-induced forces; this not only allows the preservation of gauge invariance and the conservation of momentum, but also enables velocity-dependent effects to arise naturally from the formalism. It is well known that velocity-dependent terms contribute significantly to laser cooling [4].

The standard approach to describing the effects of radiation pressure concentrates on the study of radiation-induced forces as a means of evaluating the momentum transferred during the interaction. This approach, albeit methodically standard, could lead to misleading interpretations of the formalism if the necessity for a consistent division of the motion into internal and translational components were not recognized. Such a division must be subject to the constraint that the center of mass should not introduce any additional degrees of freedom. An important consequence of the division is that the conventional dipole-approximation interaction is insufficient to describe the effects of the motion of the atomic center of mass, located at $\mathbf{R}$. This interaction, which is proportional to the atomic dipole moment $\mathbf{d}$, is loosely written as $-\mathbf{d}\cdot\mathbf{E}_{\perp}(\mathbf{R})$, although the transverse component $\mathbf{\Pi}_{\perp}$ of the canonical momentum conjugate to the vector potential $\mathbf{A}$, in the gauge that generates the interaction, is correctly identified [5] as being proportional to the transverse component $\mathbf{D}_{\perp}$ of the displacement, rather than the similar electric-field component $\mathbf{E}_{\perp}$. The transverse component of the momentum is identified as $\mathbf{E}_{\perp}$ only in the gauge corresponding to an interaction proportional to $\mathbf{p}\cdot\mathbf{A}_{\perp}$.

The adoption of the canonical procedure of quantization and the redefinition of the Hamiltonian as the quantum-mechanical time-evolution operator demand that the dynamics be evaluated within the Heisenberg formalism, consistent with the Dirac prescription of quantization. Thus, the rates of changes of the total atomic conjugate and mechanical momenta are defined, respectively, as $\dot{\mathbf{P}}=i\hbar^{-1}[H,\mathbf{P}]$ and $M\ddot{\mathbf{R}}=-\hbar^{-2}[H,[H,M\mathbf{R}]]$, where M is the total mass of the atom.

A canonical treatment of radiation pressure reveals the presence of an interaction between the canonical, gross atomic momentum $\mathbf{P}$ and a term proportional to $\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{M}}\times\mathbf{B}$, involving the magnetic field $\mathbf{B}$ and the total atomic polarization $\mathcal{P}_{\mathcal{M}}$, where the subscript $\mathcal{M}$ indicates that the full multipolar expansion of the polarization has been made. This interaction is conveniently referred to as the Röntgen interaction, since its presence is ultimately dictated by the Röntgen current, which, in turn, is a feature of the overall translational motion of any aggregate of charges [6]. The Röntgen interaction is a natural product of the formalism, and the term is needed in order to ensure energy-momentum conservation and gauge invariance of radiation-induced mechanical forces. It is important to appreciate that the Röntgen interaction is present even in the dipole-approximation regime, where it assumes a form proportional to $\mathbf{d}\times\mathbf{B}(\mathbf{R})$. The absence of the Röntgen term in most previously determined interactions comes about through the imposition of an incorrect form of the dipole approximation: one which is consistent only with a prematurely truncated version of the Lorentz force formula, and implies that the dipole only sees a parallel, spatially uniform electric field.

The role of a classical Röntgen-type interaction has recently been noted in the establishment of quasi-ionized Rydberg electronic states in crossed static $\mathbf{E}$ and $\mathbf{B}$ fields [7]. However, the term has been largely missed in previous calculations of the dynamical effects of radiation pressure, where the radiation-induced force F has been identified as the rate of change $\dot{\mathbf{P}}$ of the canonical momentum and obtained from an application of Ehrenfest's theorem in the form $\langle\mathbf{F}\rangle=\langle\nabla\{\mathbf{d}\cdot\mathbf{E}_\perp\}\rangle$. The present authors know of only one previous gross-motion calculation [8] involving the Röntgen interaction, in which a perturbative determination of the changes in momentum experienced by an atom, interacting with a single mode of laser light, indicated that the Röntgen term was responsible for momentum changes not only in the direction of the mode's wave vector $\mathbf{k}$, but also in a direction transverse to $\mathbf{k}$.

Problems of gauge and the importance of distinguishing between canonical and mechanical momenta lie at the heart of the canonical treatment of radiation-induced forces. The gauge which allows a complete separation of the internal and gross motions introduces a discrepancy between the canonical momentum $\mathbf{P}$ and the corresponding mechanical momentum $M\dot{\mathbf{R}}$. For an overall electrically neutral atom, this discrepancy is conveniently called the Röntgen momentum, since it is proportional to the Röntgen interaction, and reflects the gauge-variant nature of canonical momenta. However, the mechanical force $M\ddot{\mathbf{R}}$ is gauge invariant—a feature ensured by the presence of the Röntgen interaction. Since the force $\mathbf{F}$ is an observable, it must be identified with the gauge-invariant quantity $M\ddot{\mathbf{R}}$. In contrast, $\dot{\mathbf{P}}$ is gauge dependent. The relationship $M\ddot{\mathbf{R}}=\dot{\mathbf{P}}$ is valid only in the special case of an electrically neutral atom, when its dipole sees only a parallel, spatially uniform electric field, and only in the gauge corresponding to an interaction proportional to $\mathbf{p}\cdot\mathbf{A}_\perp$. Problems of gauge may also arise in perturbation calculations if the internal and center-of-mass components of the interaction correspond to different gauges. A sometimes-seen example of inconsistent choices occurs when the internal atom is modeled as a two-level system described by the Jaynes-Cummings Hamiltonian [9], while the atomic gross motion is described by the Hamiltonian $\mathbf{P}^2/2M$.

The layout of this paper is as follows: The Hamiltonian, which is separable into zero-order components together with internal and center-of-mass interactions, for an arbitrary number of point charges interacting with a radiation field, is canonically determined in Sec. II. The forms of the Hamiltonian and canonical momenta are further discussed in Sec. III, where, too, the electric dipole approximation is imposed. The restriction to the dipole approximation after the establishment of the zero-order Hamiltonians ensures the continuing presence of the Röntgen interaction, which is now proportional to a coupling between $\mathbf{P}$ and the dipole-approximation form $\mathbf{d}\times\mathbf{B}(\mathbf{R})$ of the Röntgen momentum. Conservation of total canonical momentum is confirmed in Sec. IV, after the specialization is made to a single mode. Also in this section, the canonical gross-motion dynamics are determined from Heisenberg's equation, with the presentation of an explicit form of $\dot{\mathbf{P}}$. A demonstration of the gauge invariance of $M\ddot{\mathbf{R}}$ follows, by expressing the mechanical force solely in terms of total electric and magnetic fields. This enables the form of the classical momentum density of an electromagnetic field to be determined, consistent with the canonical formalism. Section IV ends with some discussion of the problem of the exact form of this momentum density. The mechanical gross-motion dynamics are determined in Sec. V from Heisenberg's equation, with a calculation of $M\ddot{\mathbf{R}}$ given in terms of a single-mode field, and later specialized to the case of a two-level atom. There is a final concluding section.

II. THE MULTIPOLAR HAMILTONIAN

A single atom is here defined as an aggregate of an arbitrary number n of particles of charge e^ν and mass m^ν ($\nu=1,2,\ldots,n$), identified in the laboratory frame by the sets of canonical coordinates

$$\mathbf{q}=\{\mathbf{q}^\nu|\nu=1,2,\ldots,n\},$$
$$\mathbf{p}=\{\mathbf{p}^\nu|\nu=1,2,\ldots,n\}.$$

It is assumed that all of the particles are spinless and move at nonrelativistic speeds, consistent with the classical charge $\rho(\mathbf{r})=e^\nu\delta(\mathbf{r}-\mathbf{q}^\nu)$ and current $\mathbf{J}(\mathbf{r})=e^\nu\dot{\mathbf{q}}^\nu\delta(\mathbf{r}-\mathbf{q}^\nu)$ densities. The atom may possess a possibly nonzero net charge $e_T=\sum_\nu e^\nu$. It should be noted that throughout this paper, an Einstein summation convention is used over repeated indices, with Greek superscripts denoting a particular particle variable and Latin subscripts denoting a particular Cartesian component. It is emphasized that the summed indices are those which are found repeated on one side only of an equation, in accordance with accepted practice.

It is important to appreciate that the total atomic force $\int d^3\mathbf{r}\{\rho\mathbf{E}+\mathbf{J}\times\mathbf{B}\}$ formed from summing the Lorentz forces experienced by each charge e^ν is not particularly useful in determining those radiation forces responsible for the translational motion of the atomic center of mass relative to the laboratory observer. This is because the Lorentz force is written in terms of the total electric $\mathbf{E}$ and magnetic $\mathbf{B}$ fields, which include the intercharge Coulombic as well as the transverse radiation fields. It is much more productive to devise a formalism where the atomic dynamics are correctly separated into internal and translational components. This requires the use of the canonical procedure to obtain a Hamiltonian which is resolvable into gross and internal interactions, which, in turn, entails a canonical transformation of the minimal-coupling form of the Hamiltonian to one that expresses the atomic component of the interaction in terms of multipolar fields and the total atomic momentum $\mathbf{P}$. Some 30 years ago, Fiutak [10] obtained a completely quantum-mechanical, time-dependent unitary transformation generated by $\mathbf{r}\cdot\mathbf{A}(\mathbf{R}',t)$, where $\mathbf{R}'$ was an arbitrary coordinate origin. He did not succeed, however, in simplifying that portion of the Hamiltonian containing the magnetic field, nor, since $\mathbf{R}'$ was not the atomic center of mass $\mathbf{R}$, did he eliminate the coupling between $\mathbf{P}$ and internal atomic momenta. Fiutak's derivation is quite general. Other derivations [11–14], with some specializing to the nonrelativistic Pauli Hamiltonian [11], have expressed the transformed Hamiltonian in terms of a complete electromagnetic expansion effected by a unitary transformation of the laboratory-frame momenta using a generating function of $\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{M}}\cdot\mathbf{A}$, where $\mathcal{P}_{\mathcal{M}}$ is the total atomic polarization operator measured with respect to the center of mass $\mathbf{R}$.

The starting point for the development of our formalism is the Lagrangian

$$L^{(a)}=\frac{m^\nu}{2}\dot{\mathbf{q}}^\nu\cdot\dot{\mathbf{q}}^\nu+\int d^3\mathbf{r}\frac{\epsilon_0}{2}[(\dot{\mathbf{A}}+\nabla\varphi)^2-c^2(\nabla\times\mathbf{A})^2]+\int d^3\mathbf{r}\,\mathbf{J}\cdot\mathbf{A}-\int d^3\mathbf{r}\,\rho\varphi \tag{2.1}$$

for the above-defined atom minimally coupled to an arbitrary (a) gauge electromagnetic field. The degeneracy of (2.1) with respect to the scalar potential φ is circumvented by adopting the generalized coordinates $\mathbf{q}$, $\mathbf{A}$ as the sole independent dynamical variables [15], leading, by the usual canonical procedure, to the Hamiltonian

$$H^{(a)}=\frac{1}{2m^\nu}\{\mathbf{p}^\nu-e^\nu\mathbf{A}(\mathbf{q}^\nu)\}^2+\tfrac{1}{2}\int d^3\mathbf{r}\{\epsilon_0^{-1}\mathbf{\Pi}^2(\mathbf{r})+\mu_0^{-1}\mathbf{B}^2(\mathbf{r})\}\ , \tag{2.2}$$

with the momenta conjugate to $\mathbf{q}^\nu$ and $\mathbf{A}$ identified as

$$\mathbf{p}^\nu=m^\nu\dot{\mathbf{q}}^\nu+e^\nu\mathbf{A}(\mathbf{q}^\nu)\ , \tag{2.3a}$$

$$\mathbf{\Pi}(\mathbf{r})=-\epsilon_0\mathbf{E}(\mathbf{r})\ , \tag{2.3b}$$

respectively. There is an implicit summation over ν in (2.2) but not in (2.3a), consistent with the adopted summation convention mentioned at the beginning of this section. The conjugate coordinates obey the canonical equal-time commutators

$$[q_i^\nu,p_{i'}^{\nu'}]=i\hbar\delta_{ii'}\delta_{\nu\nu'}\ , \tag{2.4a}$$

$$[A_i(\mathbf{r}),\Pi_{i'}(\mathbf{r}')]=i\hbar\delta_{ii'}\delta(\mathbf{r}-\mathbf{r}')\ . \tag{2.4b}$$

Equations (2.2) and (2.4) are consistent within the Heisenberg formalism $\dot{O}=(i/\hbar)[H,O]$ with Maxwell's equations, the Lorentz force formula, and the equation of continuity.

To decouple the external, atomic gross motion from the internal dynamics, the phase space $\mathbf{q},\mathbf{p}$ is resolved into the component spaces $\mathbf{R}$, $\mathbf{P}$, and

$$\bar{\mathbf{q}}=\{\bar{\mathbf{q}}^\nu|\nu=1,2,\ldots,n\}\ ,$$

$$\bar{\mathbf{p}}=\{\bar{\mathbf{p}}^\nu|\nu=1,2,\ldots,n\}\ ,$$

such that

$$\mathbf{R}=\frac{m^\nu}{M}\mathbf{q}^\nu\ , \tag{2.5a}$$

$$\mathbf{P}=\sum_\nu\mathbf{p}^\nu\ , \tag{2.5b}$$

$$\bar{\mathbf{q}}^\nu=\mathbf{q}^\nu-\mathbf{R}\ , \tag{2.5c}$$

$$\bar{\mathbf{p}}^\nu=\mathbf{p}^\nu-\frac{m^\nu}{M}\mathbf{P}\ . \tag{2.5d}$$

Note that there is no summation over ν implicit in (2.5c) and (2.5d). The gross-motion dynamical variables, consisting of the center of mass (2.5a) and the total atomic conjugate momentum (2.5b), form a conjugate phase space separate from the internal phase space $\bar{\mathbf{q}},\bar{\mathbf{p}}$; therefore,

$$[R_i,P_{i'}]=i\hbar\delta_{ii'}\ , \tag{2.6a}$$

$$[\bar{q}_i^\nu,P_{i'}]=[R_i,\bar{p}_{i'}^\nu]=0\ , \tag{2.6b}$$

for all ν. The internal phase-space components (2.5c) and (2.5d) define a nonconjugate and overcomplete space from which the commutator

$$[\bar{q}_i^\mu,\bar{p}_j^\nu]=i\hbar\delta_{ij}\left\{\delta_{\mu\nu}-\frac{m^\nu}{M}\right\} \tag{2.7}$$

and equations of constraint

$$\sum_\mu\bar{\mathbf{p}}^\mu=\mathbf{0}\ , \tag{2.8a}$$

$$m^\mu\bar{\mathbf{q}}^\mu=\mathbf{0} \tag{2.8b}$$

may be constructed. A canonical commutator

$$[\bar{q}_i^\nu-\bar{q}_i^{\nu'},\bar{p}_{i'}^\nu]=i\hbar\delta_{ii'}$$

may be formed involving the position $[\bar{\mathbf{q}}^\nu-\bar{\mathbf{q}}^{\nu'}]$ of any particular particle ν relative to a different particle ν'. This is a generalization of the well-known result [16] for two particles, where (2.8a) is used to eliminate one of the internal momenta. It will be seen later that the noncanonical nature of (2.7) is harmless [17] and that the spatial overcompleteness, characterized by the equations of constraint (2.8), contributes to the elimination of unwanted interaction couplings.

In order to rewrite the Hamiltonian (2.2) in terms of the center-of-mass dynamical variables (2.5a) and (2.5b), a canonical transformation must be induced, generated by

$$\chi(\mathbf{q}^\nu)=\frac{1}{e^\nu}\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{M}}(\mathbf{r};\mathbf{q})\cdot\mathbf{A}(\mathbf{r})\ , \tag{2.9}$$

where

$$\mathcal{P}_{\mathcal{M}}(\mathbf{r};\mathbf{q})=\int_0^1 d\lambda\, e^\mu(\mathbf{q}^\mu-\mathbf{R})\delta(\mathbf{r}-\mathbf{R}-\lambda(\mathbf{q}^\mu-\mathbf{R})) \tag{2.10}$$

is the total atomic polarization written in closed form and measured with respect to the center of mass $\mathbf{R}$ defined in (2.5a). The polarization (2.10) is summed over all the charges and is therefore a function of the set $\mathbf{q}$. It is well known that the generator (2.9), with (2.10), results in the usual Power-Zienau-Woolley multipolar Hamiltonian [13,18]. The present objective, however, is to extend the formalism so as to take account of the gross motion of the charge aggregate—where $\mathbf{R}$ is a dynamical variable. The final result of the canonical transformation is to produce a Hamiltonian which, after the selection of a Coulomb (c) gauge $\nabla\cdot\mathbf{A}=0$, may be written as

$$H_{\chi}^{(c)}=H_{\bar{\mathbf{q}}}+H_{\mathbf{R}}+H_{\mathbf{r}}^{(c)}+H_i^{(c)}+\mathcal{H}_i^{(c)}\ , \tag{2.11a}$$

where

$$H_{\bar{\mathbf{q}}}=\frac{\bar{\mathbf{p}}^{\nu}\cdot\bar{\mathbf{p}}^{\nu}}{2m^{\nu}}+\frac{e^{\mu}e^{\nu}}{8\pi\epsilon_0|\bar{\mathbf{q}}^{\mu}-\bar{\mathbf{q}}^{\nu}|}\ , \tag{2.11b}$$

$$H_{\mathbf{R}}=\frac{\mathbf{P}^2}{2M}\ , \tag{2.11c}$$

$$H_{\mathbf{r}}^{(c)}=\tfrac{1}{2}\int d^3\mathbf{r}\{\epsilon_0^{-1}\mathbf{\Pi}_{\perp}^2(\mathbf{r})+\mu_0^{-1}\mathbf{B}^2(\mathbf{r})\} \tag{2.11d}$$

are the zero-order components associated with the internal atom, the center of mass, and the transverse radiation field, respectively, and

$$\begin{aligned}H_i^{(c)}=&\frac{e^{\nu}}{2m^{\nu}}\int d^3\mathbf{r}\{\bar{\mathbf{p}}^{\nu}\cdot\mathbf{\Theta}^{\nu}(\mathbf{r},\bar{\mathbf{q}}^{\nu})\times\mathbf{B}(\mathbf{r})+\mathbf{\Theta}^{\nu}(\mathbf{r},\bar{\mathbf{q}}^{\nu})\times\mathbf{B}(\mathbf{r})\cdot\bar{\mathbf{p}}^{\nu}\}\\&+\frac{1}{\epsilon_0}\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{M}}(\mathbf{r};\bar{\mathbf{q}})\cdot\mathbf{\Pi}_{\perp}(\mathbf{r})+\frac{1}{2\epsilon_0}\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{M}\perp}^2+\frac{1}{2m^{\nu}}\left\{\int d^3\mathbf{r}\,e^{\nu}\mathbf{\Theta}^{\nu}(\mathbf{r},\bar{\mathbf{q}}^{\nu})\times\mathbf{B}(\mathbf{r})\right\}^2\ ,\end{aligned} \tag{2.11e}$$

$$\begin{aligned}\mathcal{H}_i^{(c)}=&-\frac{e_T}{M}\mathbf{P}\cdot\mathbf{A}_{\perp}(\mathbf{R})+\frac{e_T^2}{2M}\mathbf{A}_{\perp}^2(\mathbf{R})+\frac{1}{2M}\int d^3\mathbf{r}\{\mathbf{P}\cdot\mathcal{P}_{\mathcal{M}}(\mathbf{r};\mathbf{q})\times\mathbf{B}(\mathbf{r})+\mathcal{P}_{\mathcal{M}}(\mathbf{r};\mathbf{q})\times\mathbf{B}(\mathbf{r})\cdot\mathbf{P}\}\\&-\frac{e_T}{M}\mathbf{A}_{\perp}(\mathbf{R})\cdot\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{M}}(\mathbf{r};\mathbf{q})\times\mathbf{B}(\mathbf{r})+\frac{1}{2M}\left\{\int d^3\mathbf{r}\mathcal{P}_{\mathcal{M}}(\mathbf{r};\mathbf{q})\times\mathbf{B}(\mathbf{r})\right\}^2\end{aligned} \tag{2.11f}$$

are the internal and center-of-mass interactions, respectively. The subscript $\perp$ in (2.11d), (2.11e), and (2.11f) indicates the transverse portion of the relevant vector, while e_T in (2.11f) denotes the total charge of the aggregate. The interactions (2.11e) and (2.11f) are written using the definitions

$$\mathcal{P}_{\mathcal{M}}(\mathbf{r};\bar{\mathbf{q}})=\int_0^1 d\lambda\, e^{\mu}\bar{\mathbf{q}}^{\mu}\delta(\mathbf{r}-\mathbf{R}-\lambda\bar{\mathbf{q}}^{\mu})\ , \tag{2.12a}$$

$$\mathbf{\Theta}^{\nu}(\mathbf{r},\bar{\mathbf{q}}^{\nu})=\int_0^1 d\lambda\,\lambda\bar{\mathbf{q}}^{\nu}\delta(\mathbf{r}-\mathbf{R}-\lambda\bar{\mathbf{q}}^{\nu})\ , \tag{2.12b}$$

where (2.12a) is the polarization (2.10) expressed explicitly in terms of the internal variables $\bar{\mathbf{q}}$.

The derivation of (2.11) is algebraically nontrivial, and the following details are important: The canonical transformation generated from (2.9), which must either be a unitary transformation of (2.2) or a gauge transformation of the potentials of (2.1), changes the momenta identifications (2.3) to those of

$$\mathbf{p}^{\nu}=m^{\nu}\dot{\mathbf{q}}^{\nu}+e^{\nu}\mathbf{A}(\mathbf{q}^{\nu})-e^{\nu}\nabla^{\nu}\chi(\mathbf{q}^{\nu})\ , \tag{2.13a}$$

$$\mathbf{\Pi}(\mathbf{r})=-\epsilon_0\mathbf{E}(\mathbf{r})-\mathcal{P}_{\mathcal{M}}(\mathbf{r};\mathbf{q})\ , \tag{2.13b}$$

where the individual-particle gradient operator $\nabla^{\nu}\equiv\partial/\partial\mathbf{q}^{\nu}$. By means of the identities

$$\nabla_i^{\nu}q_{i'}^{\nu'}=\delta_{ii'}\delta_{\nu\nu'}\ , \tag{2.14a}$$

$$\int d^3\mathbf{r}\int_0^1 d\lambda\ \mathbf{A}(\mathbf{r})\{\mathbf{q}^{\nu}-\mathbf{R}\}\cdot\nabla\delta(\mathbf{r}-\mathbf{R}-\lambda(\mathbf{q}^{\nu}-\mathbf{R}))=\mathbf{A}(\mathbf{R})-\mathbf{A}(\mathbf{q}^{\nu})\ , \tag{2.14b}$$

$$\nabla^{\nu}\delta(\mathbf{r}-\mathbf{R}-\lambda(\mathbf{q}^{\nu'}-\mathbf{R}))=\left\{-\frac{m^{\nu}}{M}-\lambda\delta_{\nu\nu'}+\lambda\frac{m^{\nu}}{M}\right\}\nabla\delta(\mathbf{r}-\mathbf{R}-\lambda(\mathbf{q}^{\nu'}-\mathbf{R}))\ , \tag{2.14c}$$

$$\begin{aligned}\int d^3\mathbf{r}\int_0^1 d\lambda\,\lambda\{A_i(\mathbf{r})(\mathbf{q}^{\nu}-\mathbf{R})\cdot\nabla-(\mathbf{q}^{\nu}-\mathbf{R})\cdot\mathbf{A}(\mathbf{r})\nabla_i\}\\\times\delta(\mathbf{r}-\mathbf{R}-\lambda(\mathbf{q}^{\nu}-\mathbf{R}))\\=\int d^3\mathbf{r}\int_0^1 d\lambda\,\lambda\{(\mathbf{q}^{\nu}-\mathbf{R})\times\mathbf{B}(\mathbf{r})\}_i\\\times\delta(\mathbf{r}-\mathbf{R}-\lambda(\mathbf{q}^{\nu}-\mathbf{R}))\ ,\end{aligned} \tag{2.14d}$$

where $\nabla\equiv\partial/\partial\mathbf{r}$, the relationship

$$\begin{aligned}e^{\nu}\nabla^{\nu}\chi(\mathbf{q}^{\nu})=&e^{\nu}\mathbf{A}(\mathbf{q}^{\nu})-\frac{m^{\nu}}{M}e_T\mathbf{A}(\mathbf{R})\\&+\int d^3\mathbf{r}\left\{e^{\nu}\mathbf{\Theta}^{\nu}(\mathbf{r},\mathbf{q}^{\nu})-\frac{m^{\nu}}{M}e^{\mu}\mathbf{\Theta}^{\mu}(\mathbf{r},\mathbf{q}^{\nu})\right.\\&\left.+\frac{m^{\nu}}{M}\mathcal{P}_{\mathcal{M}}(\mathbf{r};\mathbf{q})\right\}\times\mathbf{B}(\mathbf{r})\end{aligned} \tag{2.15}$$

may be obtained after some algebra. It should be noted that (2.15) is summed over μ but not over ν, and that $\mathbf{\Theta}^{\nu}$ and $\mathbf{\Theta}^{\mu}$ are written expressly in terms of the laboratory particle coordinates $\mathbf{q}^{\nu}$ and $\mathbf{q}^{\mu}$, implying a reexpression of (2.12b) in terms of (2.5c). The choice of a Coulomb gauge allows the resolution of zero-order atomic and radiation Hamiltonian components, with the nondynamical longitudinal field providing the Coulomb energy of the charges according to the relationship

$$(\epsilon_0/2)\int d^3\mathbf{r}\,\mathbf{E}_{\parallel}^2=e^{\mu}e^{\nu}/8\pi\epsilon_0|\bar{\mathbf{q}}^{\mu}-\bar{\mathbf{q}}^{\nu}|\ ,$$

with summations over ν and μ. The Hamiltonian (2.11) is then obtained after some algebraic manipulation and use of (2.5) and (2.8a), together with the momenta identifications

$$\bar{\mathbf{p}}^{\nu}=m^{\nu}\dot{\bar{\mathbf{q}}}^{\nu}-\int d^3\mathbf{r}\, e^{\nu}\mathbf{\Theta}^{\nu}(\mathbf{r},\bar{\mathbf{q}}^{\nu})\times\mathbf{B}(\mathbf{r}) + \frac{m^{\nu}}{M}\int d^3\mathbf{r}\, e^{\mu}\mathbf{\Theta}^{\mu}(\mathbf{r},\bar{\mathbf{q}}^{\mu})\times\mathbf{B}(\mathbf{r}) , \tag{2.16a}$$

$$\mathbf{P}=M\dot{\mathbf{R}}+e_T\mathbf{A}_{\perp}(\mathbf{R})-\int d^3\mathbf{r}\,\mathcal{P}_{M}(\mathbf{r};\mathbf{q})\times\mathbf{B}(\mathbf{r}) , \tag{2.16b}$$

$$\mathbf{\Pi}_{\perp}(\mathbf{r})=-\epsilon_0\mathbf{E}_{\perp}(\mathbf{r})-\mathcal{P}_{\mathcal{M}\perp}(\mathbf{r};\mathbf{q}) . \tag{2.16c}$$

The transverse photon fields obey the usual equal-time commutator

$$[A_{\perp i}(\mathbf{r}),\Pi_{\perp i'}(\mathbf{r}')]=i\hbar\delta_{\perp}^{ii'}(\mathbf{r}-\mathbf{r}') \tag{2.17}$$

involving the transverse δ function [19]. The details of the calculation of (2.11) reveal that all interaction terms between the polarization $\mathcal{P}_{\mathcal{M}}$, the function Θ, the total canonical momentum $\mathbf{P}$, and the internal momenta $\bar{\mathbf{p}}$ vanish. This is because of the equation of constraint (2.8a) and the fact that the polarization is written with respect to the center of mass $\mathbf{R}$, as opposed to an arbitrary coordinate origin $\mathbf{R}'$ mentioned at the beginning of this section.

The internal particle momenta (2.16a) are linear combinations of the corresponding mechanical momenta $m^{\nu}\dot{\bar{\mathbf{q}}}^{\nu}$, the function $\mathbf{\Theta}^{\mu}$, and the sum $\sum_{\mu}\mathbf{\Theta}^{\mu}$ over all the charges. They reduce to the standard Power-Zienau-Woolley [13,18] forms in the limit $M\rightarrow\infty$, becoming canonically conjugate to $\bar{\mathbf{q}}^{\nu}$. The summation of (2.16a) over ν is consistent with the equations of constraint (2.8). The gross-motion canonical momentum (2.16b) differs from the corresponding mechanical momentum $M\dot{\mathbf{R}}$ by two terms: the term proportional to the total charge e_T is consistent with the minimal coupling of that charge to the field $\mathbf{A}_{\perp}(\mathbf{R})$, while the Röntgen momentum $\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{M}}\times\mathbf{B}$ is independent of e_T. The momenta (2.16) confirm that the Hamiltonian (2.11a) may be written in the manifestly gauge-invariant form of

$$H_{\chi}^{(c)}=\frac{m^{\nu}}{2}\dot{\bar{\mathbf{q}}}^{\nu}\cdot\dot{\bar{\mathbf{q}}}^{\nu}+\frac{M}{2}\dot{\mathbf{R}}^{2}+\frac{\epsilon_0}{2}\int d^3\mathbf{r}\{\mathbf{E}_{\perp}^2(\mathbf{r})+c^2\mathbf{B}^2(\mathbf{r})\} , \tag{2.18}$$

after use of the equation of constraint (2.8a). Finally, (2.16c) confirms the identification of the momentum $-\mathbf{\Pi}_{\perp}$ with its usual form of the transverse component $\mathbf{D}_{\perp}$ of the displacement, consistent with the Power-Zienau-Woolley gauge [15].

The earlier statement that the noncanonical nature of (2.7) is harmless may be illustrated by a confirmation of (2.16a) from a determination of

$$m^{\nu}\dot{\bar{\mathbf{q}}}^{\nu}=(i/\hbar)[H_{\chi}^{(c)},m^{\nu}\bar{\mathbf{q}}^{\nu}] ,$$

involving the following nonzero components:

$$\frac{i}{\hbar}\left[\frac{\bar{\mathbf{p}}^{\nu'}\cdot\bar{\mathbf{p}}^{\nu'}}{2m^{\nu'}},m^{\nu}\bar{\mathbf{q}}^{\nu}\right]=\bar{\mathbf{p}}^{\nu} , \tag{2.19a}$$

$$\frac{i}{\hbar}\left[\frac{e^{\nu'}}{m^{\nu'}}\bar{\mathbf{p}}^{\nu'}\cdot\mathbf{\Theta}^{\nu'}(\mathbf{r},\bar{\mathbf{q}}^{\nu'}),m^{\nu}\bar{\mathbf{q}}^{\nu}\right] =e^{\nu}\mathbf{\Theta}^{\nu}(\mathbf{r},\bar{\mathbf{q}}^{\nu})-\frac{m^{\mu}}{M}e^{\mu}\mathbf{\Theta}^{\mu}(\mathbf{r},\bar{\mathbf{q}}^{\mu}) . \tag{2.19b}$$

These equations follow after the use of Eq. (2.7). It is evident from (2.19a) that the usual result of $\bar{\mathbf{p}}^{\nu}=(im^{\nu}/\hbar)[H_{\bar{q}},\bar{\mathbf{q}}^{\nu}]$, where $H_{\bar{q}}$ is the atomic zero-order Hamiltonian (2.11b), is obtained even though $\bar{\mathbf{q}}$ and $\bar{\mathbf{p}}$ are not canonical coordinates.

The zero-order component (2.11b), involving summations over ν and μ, assumes a more familiar form in the specialization to two charges $e^1=-e$, $e^2=+e$. Equation (2.8a) allows the internal momenta to be expressed in terms of $\mathbf{p}'=\bar{\mathbf{p}}^1=-\bar{\mathbf{p}}^2$ and gives

$$H_{\bar{q}}=\frac{\mathbf{p}'\cdot\mathbf{p}'}{2\mu}-\frac{e^2}{4\pi\epsilon_0|\mathbf{q}'|}+\text{infinite self-energies} \tag{2.20}$$

as the Hamiltonian, where $\mu=m^1m^2/M$ is the reduced mass and $\mathbf{q}'=\bar{\mathbf{q}}^1-\bar{\mathbf{q}}^2$. The variables $\mathbf{q}',\mathbf{p}'$ are canonical: $[q_i',p_j']=i\hbar\delta_{ij}$.

III. DISCUSSION OF THE HAMILTONIAN: THE DIPOLE APPROXIMATION

The transformed Hamiltonian (2.11) for an aggregrate of charges interacting with a radiation field exhibits the required explicit dependence on the conjugate variables (2.5a) and (2.5b) associated with the center of mass, together with a complete separation into internal (2.11e) and translational (2.11f) interactions. Equation (2.11e) corresponds to a general Power-Zienau-Woolley interaction [15], written in terms of internal momenta $\bar{\mathbf{p}}$, while (2.11f) reveals coupling between $\mathbf{P}$ and the Röntgen momentum $\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{M}}\times\mathbf{B}$, and a minimal coupling between $\mathbf{P}$ and $\mathbf{A}_{\perp}(\mathbf{R})$ governed by the total charge e_T. In the case of a neutral atom $e_T=0$, the center of mass interacts with the radiation field solely through a coupling between $\mathbf{P}$ and the Röntgen momentum, with an associated second-order self-energy given by the square of the Röntgen interaction. It will be seen that a variant of this center-of-mass coupling is present even in the dipole approximation.

The Röntgen interaction is fundamentally a characteristic of the convection current density $\mathbf{J}_R$, which results from a charge aggregate's gross motion [6]; this current is extra to the aggregrate's internal (bound) currents and to any free currents present due to a nonzero overall charge. The total current density is the sum of these effects, and it assumes the most general form of

$$\mathbf{J}(\mathbf{r})=\frac{e_T}{2}\{\dot{\mathbf{R}}\delta(\mathbf{r}-\mathbf{R})+\delta(\mathbf{r}-\mathbf{R})\dot{\mathbf{R}}\}+\mathbf{J}'(\mathbf{r}) , \tag{3.1}$$

where the current density

$$\mathbf{J}'(\mathbf{r})=\mathbf{J}_R(\mathbf{r})+\dot{\mathcal{P}}_{\mathcal{M}}(\mathbf{r};\bar{\mathbf{q}})+\nabla\times\mathbf{M}(\mathbf{r};\bar{\mathbf{q}}) \tag{3.2a}$$

for a neutral charge aggregrate $e_T=0$ is the sum of the Röntgen current

$$\mathbf{J}_R(\mathbf{r})=\tfrac{1}{2}\nabla\times\{\mathcal{P}_{\mathcal{M}}(\mathbf{r};\bar{\mathbf{q}})\times\dot{\mathbf{R}}-\dot{\mathbf{R}}\times\mathcal{P}_{\mathcal{M}}(\mathbf{r};\bar{\mathbf{q}})\} \tag{3.2b}$$

and the usual electric polarization and magnetization currents, with

$$\mathbf{M}(\mathbf{r};\bar{\mathbf{q}})=\tfrac{1}{2}\int_0^1 d\lambda\,\lambda e^{\mu}\{\bar{\mathbf{q}}^{\mu}\times\dot{\bar{\mathbf{q}}}^{\mu}-\dot{\bar{\mathbf{q}}}^{\mu}\times\bar{\mathbf{q}}^{\mu}\}\delta(\mathbf{r}-\mathbf{R}-\lambda\bar{\mathbf{q}}^{\mu}) \tag{3.2c}$$

as the magnetization operator. Equation (3.1) is an identity, following from the Dirac δ-function properties of the closed forms of $\mathcal{P}_{\mathcal{M}}$ and $\mathbf{M}$ [20] independently of any canonical formalism. However, a verification of (3.1) from a determination within the Heisenberg formalism requires the inclusion of all the terms in the Hamiltonian (2.11); in particular, $\mathbf{J}_R$ would not appear if the Röntgen interaction were absent from (2.11f).

The adoption of the dipole approximation has the effect of truncating the expansions (2.10) and (2.12) to their leading terms of

$$\mathcal{P}_{\mathcal{D}}=\mathbf{d}\delta(\mathbf{r}-\mathbf{R}) , \tag{3.3a}$$

$$\Theta_{\mathcal{D}}^{\nu}=\tfrac{1}{2}\bar{\mathbf{q}}^{\nu}\delta(\mathbf{r}-\mathbf{R}) , \tag{3.3b}$$

where $\mathbf{d}=e^{\mu}\bar{\mathbf{q}}^{\mu}$ is the dipole moment. This, in turn, rewrites the interactions (2.11e) and (2.11f) as

$$H_i^{(c)}=\epsilon_0^{-1}\mathbf{d}\cdot\mathbf{\Pi}_{\perp}(\mathbf{R})+\frac{1}{2\epsilon_0}\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{D}\perp}^2(\mathbf{r}) , \tag{3.4a}$$

$$\begin{aligned}\mathcal{H}_i^{(c)}=\frac{1}{2M}[&\{\mathbf{P}-e_T\mathbf{A}_{\perp}(\mathbf{R})\}\cdot\mathbf{d}\times\mathbf{B}(\mathbf{R})\\&+\mathbf{d}\times\mathbf{B}(\mathbf{R})\cdot\{\mathbf{P}-e_T\mathbf{A}_{\perp}(\mathbf{R})\}]\\&-\frac{e_T}{2M}\{\mathbf{P}\cdot\mathbf{A}_{\perp}(\mathbf{R})+\mathbf{A}_{\perp}(\mathbf{R})\cdot\mathbf{P}\}\\&+\frac{e_T^2}{2M}\mathbf{A}_{\perp}^2(\mathbf{R})+\frac{1}{2M}\{\mathbf{d}\times\mathbf{B}(\mathbf{R})\}^2 ,\end{aligned} \tag{3.4b}$$

respectively. It should be noted that the magnetic dipole and diamagnetic interactions, formed from (3.3b), have been omitted from (3.4a). This is justified on the grounds that the former interaction is smaller than the first term of (3.4a) by the order of the fine-structure constant, and the latter is second order in photon destruction and creation operators [21]. The last term of (3.4a) and the last two terms of (3.4b) are also of second order and are similarly ignorable. The last term of (3.4a) contains the transverse component of (3.3a), such that

$$\mathcal{P}_{\mathcal{D}\perp i}(\mathbf{r})=d_{i'}\delta_{\perp}^{i'i}(\mathbf{r}-\mathbf{R}) ,$$

and is important in the calculations of the Lamb shift and van der Waals' forces. Finally, the canonical momenta are identified as

$$\bar{\mathbf{p}}^{\nu}=m^{\nu}\dot{\bar{\mathbf{q}}}^{\nu} , \tag{3.5a}$$

$$\mathbf{P}=M\dot{\mathbf{R}}+e_T\mathbf{A}_{\perp}(\mathbf{R})-\mathbf{d}\times\mathbf{B}(\mathbf{R}) , \tag{3.5b}$$

$$\mathbf{\Pi}_{\perp}(\mathbf{r})=-\epsilon_0\mathbf{E}_{\perp}(\mathbf{r})-\mathcal{P}_{\mathcal{D}\perp}(\mathbf{r};\bar{\mathbf{q}}) \tag{3.5c}$$

in the dipole-approximation regime.

It may be seen that the internal momenta (3.5a) are solely mechanical in the dipole approximation. On the other hand, the gross-motion particle momentum $\mathbf{P}$ retains a nonmechanical component, even in the case of a neutral atom, and the Röntgen momentum remains present in the form of $\mathbf{d}\times\mathbf{B}(\mathbf{R})$. This is contrary to the naive approach of introducing the dipole approximation, before applying the canonical transformation, by rewriting the vector potential in (2.2) as $\mathbf{A}(\mathbf{R})$ and the generator (2.9) as $(1/e^{\nu})\mathbf{d}\cdot\mathbf{A}(\mathbf{R})$; this results in the loss of the Röntgen interaction from the Hamiltonian and the loss of the differentiation between the canonical and mechanical gross-motion momenta.

If one were to write naively $\mathbf{A}_{\perp}(\mathbf{q}^{\nu})$ as $\mathbf{A}_{\perp}(\mathbf{R})$, consistent with $\bar{\mathbf{q}}^{\nu}\cdot\mathbf{k}\ll 0$ for all ν and $\mathbf{k}$, then the Hamiltonian (2.2), in the absence of any canonical transformation and in conjunction with (2.5) and (2.8a), might, formally, be reexpressed in a Coulomb gauge as

$$H^{(c)}=H_{\bar{\mathbf{q}}}+H_{\mathbf{R}}+H_{\mathbf{r}}^{(c)}+H_{\mathrm{int}}^{(c)} , \tag{3.6}$$

where the first three terms of (3.6) remain as (2.11b)–(2.11d) but where the total interaction is expressed in the $\mathbf{p}\cdot\mathbf{A}_{\perp}$ gauge as

$$H_{\mathrm{int}}^{(c)}=\frac{e^{\nu}}{m^{\nu}}\bar{\mathbf{p}}^{\nu}\cdot\mathbf{A}_{\perp}(\mathbf{R})+\frac{e^{\nu}e^{\nu}}{2m^{\nu}}\mathbf{A}_{\perp}^2(\mathbf{R})-\frac{e_T}{M}\mathbf{P}\cdot\mathbf{A}_{\perp}(\mathbf{R}) . \tag{3.7}$$

The momenta of (3.6) may be identified as

$$\bar{\mathbf{p}}^{\nu}=m^{\nu}\dot{\bar{\mathbf{q}}}^{\nu}+\left\{e^{\nu}-\frac{e_Tm^{\nu}}{M}\right\}\mathbf{A}_{\perp}(\mathbf{R}) , \tag{3.8a}$$

$$\mathbf{P}=M\dot{\mathbf{R}}+e_T\mathbf{A}_{\perp}(\mathbf{R}) , \tag{3.8b}$$

$$\mathbf{\Pi}_{\perp}(\mathbf{r})=-\epsilon_0\mathbf{E}_{\perp}(\mathbf{r}) . \tag{3.8c}$$

As mentioned in Sec. I, the canonical and mechanical gross momenta are now equivalent in the case of a neutral atom. However, the form of the electromagnetic force experienced by each charge e^{ν} consistent with (3.6) is not given by the Lorentz force formula, since the spatial dependence of the force on the positions of e^{ν} has been lost in the degeneration to a uniform $\mathbf{R}$ dependence. Thus the dipole $\mathbf{d}$ sits in a spatially uniform field, which, in the case of a Coulomb gauge, implies that the direction of $\mathbf{d}$ is parallel to the electric field $\mathbf{E}_{\perp}$. Such an orientation is often assumed, as in, for example, the Einstein-Hopf model [22] for thermodynamical equilibrium between a dipole and an electromagnetic field, and corresponds to modeling the atom as an infinitely massive oscillator. A proper description of radiation-induced atomic gross motion, however, where Doppler terms are present in the formalism and with no implicit assumptions about the orientation of $\mathbf{d}$, must be determined by making the dipole approximation in its correct form of (3.3) *after* the application of the canonical transformation generated by (2.9).

IV. CANONICAL GROSS-MOTION DYNAMICS

Now that the canonical framework in the form of the Hamiltonian (2.11) and its electric dipole approximation interaction (3.4) has been established, the present and following sections are devoted to its use in determining the atomic gross-motion dynamics. The rate of change of the total canonical momentum is calculated in Sec. IV, allowing that of the mechanical momentum to be expressed in a manifestly gauge-invariant form involving the total electric and magnetic fields. In Sec. V, the mechanical force $M\ddot{\mathbf{R}}$ is reexpressed in terms of a transverse radiation mode, where, too, the specialization to an atomic two-level system is considered. In both sections, calculations are made on the bases of the dipole approximation and a single-mode $\mathbf{k}=(\omega/c)\hat{\mathbf{k}}$, and the notation of a tilde is adopted to denote some particular quantities measured with respect to the single mode.

The dipole-approximation, single-mode Hamiltonian $\widetilde{H}_\chi^{(c)}$ may be constructed from the zero-order components (2.11b)–(2.11d) and the interaction (3.4) by replacing the radiation fields with the single-mode quantities

$$\widetilde{\mathbf{A}}_\perp(\mathbf{r})=\left[\frac{\hbar}{2\epsilon_0\omega V}\right]^{1/2}\hat{\mathbf{u}}\{a\exp(i\mathbf{k}\cdot\mathbf{r})+a^\dagger\exp(-i\mathbf{k}\cdot\mathbf{r})\}\ , \tag{4.1a}$$

$$\widetilde{\mathbf{\Pi}}_\perp(\mathbf{r})=-i\left[\frac{\hbar\epsilon_0\omega}{2V}\right]^{1/2}\hat{\mathbf{u}}\{a\exp(i\mathbf{k}\cdot\mathbf{r})-a^\dagger\exp(-i\mathbf{k}\cdot\mathbf{r})\}\ , \tag{4.1b}$$

with $\widetilde{\mathbf{B}}(\mathbf{r})=\nabla\times\widetilde{\mathbf{A}}_\perp(\mathbf{r})$. Equations (4.1) are written in terms of the usual single-mode Bosonic operators $a,a^\dagger$; and the polarization vector $\hat{\mathbf{u}}$ is assumed to be real. The cavity quantization volume V normally cancels out in the calculations of observables, and may be allowed therefore to tend to infinity. It should perhaps be mentioned that the commutators formed from the single-mode fields by the use of $[a,a^\dagger]=1$ are not equivalent to their usual multimode forms—for example, the commutator

$$[\widetilde{A}_{\perp i}(\mathbf{r}),\widetilde{\Pi}_{\perp i'}(\mathbf{r}')]=(i\hbar/V)\hat{u}_i\hat{u}_{i'}\cos[\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')]$$

should be compared with (2.17).

The determination of $\dot{\mathbf{P}}=(i/\hbar)[\widetilde{H}_\chi^{(c)},\mathbf{P}]$ follows straightforwardly—albeit tediously—using the commutators (2.6a) and $[a,a^\dagger]=1$. The result is

$$\dot{\mathbf{P}}=-\omega\mathbf{k}\mathbf{d}\cdot\widetilde{\mathbf{A}}_\perp(\mathbf{R})+\frac{\mathbf{k}}{2}\{\dot{\mathbf{R}}\cdot\mathbf{d}\times[\mathbf{k}\times\widetilde{\mathbf{A}}_\perp(\mathbf{R})]+\mathbf{d}\times[\mathbf{k}\times\widetilde{\mathbf{A}}_\perp(\mathbf{R})]\cdot\dot{\mathbf{R}}\}-\frac{e_T}{2\omega\epsilon_0}\mathbf{k}\{\dot{\mathbf{R}}\cdot\widetilde{\mathbf{\Pi}}_\perp(\mathbf{R})+\widetilde{\mathbf{\Pi}}_\perp(\mathbf{R})\cdot\dot{\mathbf{R}}\}\ . \tag{4.2}$$

The first term of (4.2) arises from the first term in the single-mode form of the internal interaction (3.4a), while the second term of (4.2) is due to the presence of the Röntgen interaction term in the single-mode form of the center-of-mass interaction (3.4b). The final term of (4.2) obviously vanishes in the case of a neutral atom. The rate of change of canonical momentum is in the direction $\hat{\mathbf{k}}$ of the mode vector, and it is consistent with the conservation of momentum in the form

$$\frac{d}{dt}\left[\mathbf{P}+\int d^3\mathbf{r}\,\widetilde{\mathbf{G}}\right]=\mathbf{0}\ , \tag{4.3}$$

where

$$\widetilde{\mathbf{G}}=\tfrac{1}{2}\{\widetilde{\mathbf{B}}(\mathbf{r})\times\widetilde{\mathbf{\Pi}}_\perp(\mathbf{r})-\widetilde{\mathbf{\Pi}}_\perp(\mathbf{r})\times\widetilde{\mathbf{B}}(\mathbf{r})\} \tag{4.4}$$

is the single-mode momentum-density component of the energy-momentum tensor parallel to $\hat{\mathbf{k}}$. Substitutions of single-mode forms of (3.5b) and (3.5c) into (4.3) allow the mechanical force operator for a single mode to be written in terms of single-mode electric $\widetilde{\mathbf{E}}_\perp$ and magnetic $\widetilde{\mathbf{B}}$ fields as

$$M\ddot{\mathbf{R}}=e_T\widetilde{\mathbf{E}}_\perp(\mathbf{R})-\frac{\epsilon_0}{2}\frac{d}{dt}\int d^3\mathbf{r}\{\widetilde{\mathbf{E}}_\perp(\mathbf{r})\times\widetilde{\mathbf{B}}(\mathbf{r})-\widetilde{\mathbf{B}}(\mathbf{r})\times\widetilde{\mathbf{E}}_\perp(\mathbf{r})\}\ . \tag{4.5}$$

Equation (4.5) is in a manifestly gauge-invariant form: a generalization to multimode $\mathbf{E}_\perp$ and $\mathbf{B}$ fields is trivial. A similar result would be obtained had (4.3) been determined in the absence of the dipole approximation. Inherent in the canonical formalism is the assumption that all fields vanish at infinity, and this, in conjunction with the fact that spatial integration is here taken throughout all space, means that no surface term is present in (4.5). It is important to appreciate that the ability to express $M\ddot{\mathbf{R}}$ in terms of gauge-invariant fields comes about because of the presence of the Röntgen momentum term in the identification of $\mathbf{P}$. This term, which is proportional to the transverse component of the polarization, is removed by a similar term, but of opposite sign, brought to the energy-momentum tensor through the identification of $\mathbf{\Pi}_\perp$ as the transverse component $-\mathbf{D}_\perp$ of the displacement.

This latter identification is also responsible for the multimode generalization of (4.4) being consistent with the Minkowski [23] form $\mathbf{D}_\perp\times\mathbf{B}$ of the classical momentum density in a Coulomb gauge electromagnetic field in a dielectric medium. It may seem possible that a claim of consistency with the Abraham [24] form $\epsilon_0\mathbf{E}_\perp\times\mathbf{B}$ of this density would come about by adopting the $\mathbf{p}\cdot\mathbf{A}_\perp$ gauge identification (3.8c) of the conjugate momentum in terms of the electric field. However, as was seen at the end of Sec. III, this assumes that $\mathbf{d}$ lies exactly parallel to the direction of an electric field which is uniform across the dipole [25]. Even here, the formalism involves subtleties which are not readily apparent. A canonical transformation of the Hamiltonian (2.2) induced by the generalization $\chi'=\zeta\chi$ of (2.9), where ζ is an arbitrary parameter, produces a ζ-dependent total interaction and ζ-dependent conjugate momenta [26]. This interaction is formally separable into internal and center-of-mass components either in the gauge corresponding to $\zeta=1$, resulting in (2.11e) and (2.11f), or, for general ζ, if it assumed that every charge sees the vector potential as $\mathbf{A}_\perp(\mathbf{R})$. Momentum is still conserved in the latter case, which, as was seen in Sec. III, corresponds to $\mathbf{d}$ being exactly parallel to $\mathbf{E}_\perp$, and the gauge-invariant form (4.5) of the mechanical force may be obtained for a single mode, but all at the expense of a ζ-dependent energy-momentum tensor. Consequently, the introduction of a prematurely truncated form of the dipole approximation, in replacing $\mathbf{A}_\perp(\mathbf{q}^\nu)$ by $\mathbf{A}_\perp(\mathbf{R})$, implies a gauge-dependent classical momentum density—which assumes the Abraham form with the choice of $\zeta=0$ and the Minkowski form when $\zeta=1$. Thus, self-consistency is only maintained if the interaction is separated by putting $\zeta=1$ and introducing the dipole approximation in the correct manner.

It should be noted that the formalism's support for the Minkowski momentum density comes from a rigorous, canonical argument independent of any particular physical model. In general, as can be seen from (4.5), the total mechanical momentum of the atom and electromagnetic field is not conserved. Only in the special case of $e_T=0$ is it possible to claim that $\epsilon_0\mathbf{E}_\perp\times\mathbf{B}$ represents the mechanical momentum density of a multimode classical electromagnetic field. Equation (4.3) is the sole expression of momentum conservation, remaining valid for all e_T. This is in keeping with Noether's principle expressed in terms of the invariance of the Lagrangian (2.1) under infinitesimal transformations of the generalized coordinates $\mathbf{q}^\nu$ and $\mathbf{A}(\mathbf{q}^\nu)$.

In no way is it claimed that the arguments presented in this section represent the resolution of the Minkowski-Abraham controversy [27] on the correct form of the electromagnetic momentum density in a material medium. However, a first attempt has been made to examine the issue from a quantized canonical perspective, in which the matter and fields enter the formalism on an equal footing, and, as such, the treatment is consistent with the microscopic physics of particles. In contrast to previous investigations, which have all depended upon classical, continuum determinations of the pressure on an imaginary surface drawn within a region of an electromagnetic field, the present authors have given a more fundamental study involving calculations, from a quantum electrodynamical Hamiltonian, of the rates of change of the canonical and mechanical momenta operators for an aggregate of an arbitrary number of microscopic charges interacting with the field.

So far, there has been no mention of pseudomomentum or crystal momentum. Translational invariance of the whole dynamical system, which is composed of charge particles and the electromagnetic field, gives rise to the conservation of total momentum, and, ultimately, to Eq. (4.3). However, translation invariance of the charge aggregate alone, in the absence of the electromagnetic field, gives rise to what has been defined [28] as the pseudomomentum density $\mathbf{K}$. There is no direct equivalence to $\mathbf{K}$ in this paper, since, as has been shown, changes in gross motion stem from a coupling between the charges and the field, and the subsystems should not be considered in isolation. However, for an overall electrically neutral charge aggregate, the pseudomomentum may be thought of as $-\mathbf{P}\delta(\mathbf{r}-\mathbf{R})$, if the Röntgen momentum $\mathbf{d}\times\mathbf{B}(\mathbf{R})$ is ignored. In this form $\mathbf{K}$ is approximately equal to $\mathbf{G}$, as shown elsewhere [29] and, of course, the distinction between canonical and mechanical momenta has been lost. It is interesting to note that the conditions under which it is generally believed that conservation of pseudomomentum holds are compatible with ignoring the Röntgen term.

Finally, it is confirmed that the formalism developed in this paper, as represented by Eqs. (3.5b), (4.3), and (4.5), is consistent with a linear electric susceptibility for a charge aggregate of general e_T given by its relative permittivity minus 1.

V. MECHANICAL GROSS-MOTION DYNAMICS

To be able to perform quantum-mechanical calculations of gross-motion mechanical forces, it is more useful to express (4.5) in terms of the radiation mode, and, therefore, in terms of photon annihilation and creation operators. This involves a direct determination of

$$M\ddot{\mathbf{R}}=-\hbar^{-2}[\tilde{H}_\chi^{(c)},[\tilde{H}_\chi^{(c)},M\mathbf{R}]] \;.$$

After some algebra, making use of (4.2) and the relationships

$$\dot{\tilde{\mathbf{A}}}_\perp(\mathbf{R})=\epsilon_0^{-1}\tilde{\mathbf{\Pi}}_\perp(\mathbf{R})-\frac{1}{2\epsilon_0\omega}[\dot{\mathbf{R}}\cdot\mathbf{k}\tilde{\mathbf{\Pi}}_\perp(\mathbf{R})+\tilde{\mathbf{\Pi}}_\perp(\mathbf{R})\mathbf{k}\cdot\dot{\mathbf{R}}] \;, \tag{5.1a}$$

$$\dot{\tilde{\mathbf{B}}}(\mathbf{R})=\epsilon_0^{-1}\nabla\times\tilde{\mathbf{\Pi}}_\perp(\mathbf{R})-\tfrac{1}{2}[\dot{\mathbf{R}}\cdot\mathbf{k}\{\mathbf{k}\times\tilde{\mathbf{A}}_\perp(\mathbf{R})\}+\{\mathbf{k}\times\tilde{\mathbf{A}}_\perp(\mathbf{R})\}\mathbf{k}\cdot\dot{\mathbf{R}}] \;, \tag{5.1b}$$

as well as the identity

$$\epsilon_0\omega\mathbf{X}\times\tilde{\mathbf{B}}=-\mathbf{k}(\mathbf{X}\cdot\tilde{\mathbf{\Pi}}_\perp)+(\mathbf{X}\cdot\mathbf{k})\tilde{\mathbf{\Pi}}_\perp \tag{5.2}$$

for any vector $\mathbf{X}$, it is found that

$$\begin{aligned}M\ddot{\mathbf{R}}=&-\omega(\mathbf{d}\cdot\mathbf{k})\tilde{\mathbf{A}}_\perp(\mathbf{R})-\tfrac{1}{2}(\mathbf{d}\cdot\mathbf{k})[\dot{\mathbf{R}}\times\{\mathbf{k}\times\tilde{\mathbf{A}}_\perp(\mathbf{R})\}-\{\mathbf{k}\times\tilde{\mathbf{A}}_\perp(\mathbf{R})\}\times\dot{\mathbf{R}}]\\&+\dot{\mathbf{d}}\times\tilde{\mathbf{B}}(\mathbf{R})-\frac{e_T}{\epsilon_0}\tilde{\mathbf{\Pi}}_\perp(\mathbf{R})\\&+\frac{e_T}{2}[\dot{\mathbf{R}}\times\tilde{\mathbf{B}}(\mathbf{R})-\tilde{\mathbf{B}}(\mathbf{R})\times\dot{\mathbf{R}}] \;.\end{aligned} \tag{5.3}$$

The operator (5.3), which is exact to within the electric-dipole approximation, forms one of the main results of the present paper. It expresses the quantum-mechanical force, induced by radiation pressure from a single mode, on an aggregate of charges modeled as a composite comprising a point charge e_T and a dipole $\mathbf{d}$. The operator $\dot{\mathbf{R}}$ in (5.3) may be replaced by $\mathbf{P}/M$ from a single-mode variant of (3.5b) if second-order terms in photon creation and annihilation operators are ignored. Thus, the velocity-dependent terms occur as a natural result of the formalism and do not have to be added "by hand."

The single-mode force operator (5.3) may be compared with a multimode classical force

$$[M\ddot{\mathbf{R}}]_{\text{classical}}=(\mathbf{d}\cdot\nabla)\mathbf{E}+(\mathbf{d}\cdot\nabla)\dot{\mathbf{R}}\times\mathbf{B}+\dot{\mathbf{d}}\times\mathbf{B} \;, \tag{5.4}$$

obtained elsewhere [30], for a neutral atom. A rearrangement of (5.4) from Maxwell's equation $\dot{\mathbf{B}}=-\nabla\times\mathbf{E}$ gives

$$[M\ddot{\mathbf{R}}]_{\text{classical}}=\nabla\{(\mathbf{E}+\dot{\mathbf{R}}\times\mathbf{B})\cdot\mathbf{d}\}+\frac{d}{dt}(\mathbf{d}\times\mathbf{B}) . \quad (5.5)$$

If the total temporal derivative in (5.5) is ignored as having no net effect on atomic gross-motion dynamics over a long time period and if terms in $\dot{\mathbf{R}}$ are ignored as being of the order of c^{-1}, then the remaining force of (5.5) may be identified as that obtained by the application of Ehrenfest's theorem to the $\mathbf{d}\cdot\mathbf{E}_{\perp}$ interaction.

A useful specialization of (5.3) involves modeling the atom as a neutral $e_T=0$ two-level system, with the eigenenergy of the ground state $|g\rangle$ taken as zero and that of the excited state $|e\rangle$ as $\hbar\omega_0$, such that $H_{\bar{q}}=\hbar\omega_0|e\rangle\langle e|$ is its zero-order Hamiltonian. The dipole moment and its rate of change may be written in terms of the usual two-level raising $\pi^{\dagger}=|e\rangle\langle g|$ and lowering $\pi=|g\rangle\langle e|$ dyadics, viz.,

$$\mathbf{d}=-e(\pi+\pi^{\dagger})\langle g|\mathcal{D}|e\rangle , \quad (5.6a)$$

$$\dot{\mathbf{d}}=ie\omega_0(\pi-\pi^{\dagger})\langle g|\mathcal{D}|e\rangle . \quad (5.6b)$$

The vector $\mathcal{D}=\sum_{\alpha}\{\bar{\mathbf{q}}^{\alpha}_{(-)}-\bar{\mathbf{q}}_{(+)}\}$ is the sum over all α electrons of the differences between their positions $\bar{\mathbf{q}}^{\alpha}_{(-)}$ and the position $\bar{\mathbf{q}}_{(+)}$ of the nucleus, measured with respect to $\mathbf{R}$. It is assumed that the wave functions corresponding to the atomic ground and excited states are real [31] and, therefore, $\langle g|\mathcal{D}|e\rangle=\langle e|\mathcal{D}|g\rangle$. With $e_T=0$, a substitution of (5.6) into (5.3) reveals that the force

$$M\ddot{\mathbf{R}}=F_1\hat{\mathbf{k}}+F_2\hat{\mathbf{u}} \quad (5.7a)$$

experienced by a two-level atom is composed of a component

$$F_1=\hbar|\mathbf{k}|g^{(\hat{\mathbf{u}})}\{W-W'\}\frac{\omega_0}{\omega^{1/2}}+|\mathbf{k}|^2g^{(\hat{\mathbf{k}})}\{W+W'\}\frac{\hbar}{\omega^{1/2}}\frac{\mathbf{P}\cdot\hat{\mathbf{u}}}{M} \quad (5.7b)$$

in the direction $\hat{\mathbf{k}}$ of the wave vector and a component

$$F_2=\hbar|\mathbf{k}|g^{(\hat{\mathbf{k}})}\left\{W\left[1-\frac{\omega_0}{\omega}\right]+W'\left[1+\frac{\omega_0}{\omega}\right]\right\}\omega^{1/2}-|\mathbf{k}|^2g^{(\hat{\mathbf{k}})}\{W+W'\}\frac{\hbar}{\omega^{1/2}}\frac{\mathbf{P}\cdot\hat{\mathbf{k}}}{M}-|\mathbf{k}|^3g^{(\hat{\mathbf{k}})}\{W+W'\}\frac{\hbar^2}{2\omega^{1/2}M} \quad (5.7c)$$

in the direction $\hat{\mathbf{u}}$ of the polarization. In (5.7), the so-called rotating terms, that is, those ladder operators' products which are not discarded by an application of the rotating-wave approximation [32], are grouped with their corresponding spatial dependence as the operator

$$W=\exp(-i\mathbf{k}\cdot\mathbf{R})a^{\dagger}\pi+\pi^{\dagger}a\exp(i\mathbf{k}\cdot\mathbf{R}) , \quad (5.8a)$$

while the counterrotating terms are similarly grouped as

$$W'=\exp(-i\mathbf{k}\cdot\mathbf{R})a^{\dagger}\pi^{\dagger}+\pi a\exp(i\mathbf{k}\cdot\mathbf{R}) . \quad (5.8b)$$

The parameters

$$g^{(\hat{\mathbf{k}})}=e\left[\frac{1}{2\epsilon_0\hbar V}\right]^{1/2}\langle g|\mathcal{D}\cdot\hat{\mathbf{k}}|e\rangle , \quad (5.9a)$$

$$g^{(\hat{\mathbf{u}})}=e\left[\frac{1}{2\epsilon_0\hbar V}\right]^{1/2}\langle g|\mathcal{D}\cdot\hat{\mathbf{u}}|e\rangle \quad (5.9b)$$

express the orientation of the dipole.

The first and third terms of (5.3) give rise to the first terms of (5.7b) and (5.7c). The dominant resonance effect of the first term of (5.7b), ignoring the counterrotating terms (5.8b), is provided by the component of $\dot{\mathbf{d}}\times\tilde{\mathbf{B}}(\mathbf{R})$ in the $\hat{\mathbf{k}}$ direction, that is, by

$$-(|\mathbf{k}|/\omega\epsilon_0)\dot{\mathbf{d}}\times\tilde{\mathbf{\Pi}}_{\perp}(\mathbf{R}) .$$

This is consistent with a determination of the radiation-induced force from an application of Ehrenfest's theorem, providing the dipole is driven in the manner of a forced harmonic oscillator by a local electric field indistinguishable from $-\epsilon_0^{-1}\tilde{\mathbf{\Pi}}_{\perp}$, consistent with the Einstein-Hopf model [22]. However, the orientation of the dipole cannot be taken necessarily as being along $\hat{\mathbf{u}}$ for a full description of the atomic gross motion to be made. In particular, such an orientation destroys the velocity-dependent terms of (5.7), which have arisen naturally from the formalism. The final component of (5.7c) represents a recoil effect and arises directly from the action of $\mathbf{P}$, in the form of a differentiation with respect to $\mathbf{R}$, on the Röntgen momentum $\mathbf{d}\times\tilde{\mathbf{B}}(\mathbf{R})$.

One might naively suppose that the component $F_2\hat{\mathbf{u}}$ of (5.7a) in the direction of the photon polarization $\hat{\mathbf{u}}$ would cycle-average to zero. However, it must be remembered that (4.12) is not a classical force, but a dynamical operator. Thus, a determination of the expectation value $\langle M\ddot{\mathbf{R}}\rangle$ over a time interval τ requires that the operators at time t of (4.12) be rewritten in terms of corresponding operators at time zero. If this is done and the steady-state $\tau\to\infty$ solution sought, then the transverse physical momentum change is nonzero and acts in a direction dictated by the orientation of the dipole moment [33].

VI. CONCLUSIONS

The canonical approach to the calculations of atomic gross motion due to radiation pressure has been presented in this paper. A canonical transformation generated by (2.9) enables the minimal-coupling Hamiltonian (2.2) to be expressed in a form (2.11), which resolves the internal and gross motions of an aggregrate composed of an arbitrary number of charges. Such a resolution makes possible a rigorous treatment of the gross motion within the Heisenberg formalism, revealing, through the presence in the interaction (2.11f) of the Röntgen momentum $\int d^3\mathbf{r}\,\mathcal{P}_{\mathcal{M}}\times\mathbf{B}$, the nonequivalence (2.16b) of the canonical $\mathbf{P}$ and mechanical $M\dot{\mathbf{R}}$ momenta, even in the case of an electrically neutral aggregate.

The specialization in Sec. III to the correct form of the dipole approximation truncates the Röntgen momentum term to $\mathbf{d}\times\mathbf{B}(\mathbf{R})$, with $\mathbf{P}$ now identified by (3.5d). This is in contrast to the usually seen approach of imposing a prematurely truncated form of the dipole approximation by writing $\mathbf{A}_\perp(\mathbf{q}^\nu)$ as $\mathbf{A}_\perp(\mathbf{R})$ for all ν, resulting in the absence of any Röntgen-type interaction. The total momentum of the atom and the radiation field, specialized to a single mode, is shown in Sec. IV to be a constant of motion (4.3), through the definition (4.4) of the electromagnetic momentum density parallel to $\hat{\mathbf{k}}$. This enables an explicit demonstration of gauge invariance of the mechanical force to be made (4.5), and a demonstration of consistency of the canonical formulation of photon pressure with the Minkowski form of momentum density.

The determination of the single-mode physical force operator (5.3) is exact to within the dipole approximation; it reveals in a manner consistent with the formalism the forms of the velocity-dependent effects and the presence of components along the directions of the wave and the polarization vectors. Work is in progress, stemming from the specialization (5.7) of the force operator to the case of a two-level atom, on a canonical examination of the trapping forces generated by counterpropagating beams—currently, an area of great interest. In particular, the time evolution of the expectation value of (5.7c) is shown to be significant in certain special cases involving one-dimensional optical molasses, as, for example, when the direction of the atomic beam is almost at right angles to the radiation beams [33].

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