

Multipole expansions and intense fields

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In the context of two-body bound-state systems subjected to a plane-wave electromagnetic field, it is shown that high field intensity introduces a distinction between long-wavelength approximation and electric dipole approximation. This distinction is gauge dependent, since it is absent in Coulomb gauge, whereas in “completed” gauges of Göppert-Mayer type the presence of high field intensity makes electric quadrupole and magnetic dipole terms of importance equal to electric dipole at long wavelengths. Another consequence of high field intensity is that multipole expansions lose their utility in view of the equivalent importance of a number of low-order multipole terms and the appearance of large-magnitude terms which defy multipole categorization. This loss of the multipole expansion is gauge independent. Also gauge independent is another related consequence of high field intensity, which is the intimate coupling of center-of-mass and relative coordinate motions in a two-body system.

I. INTRODUCTION

Very intense electromagnetic plane-wave fields are responsible for a variety of physical effects that are qualitatively different from those familiar in electrodynamics at ordinary intensities. The specific intense field phenomena examined in this paper are the loss of utility of the usual multipole expansion of the electromagnetic field, and the intimate coupling that the field introduces between center-of-mass (c.m.) and relative motions in a two-body system of charged particles.

The work below is carried out in the context of the two-body nonrelativistic Schrödinger equation in semiclassical electrodynamics. One feature of very intense fields is that charged particles can acquire relativistic velocities in a low-energy problem that is otherwise entirely nonrelativistic. Nevertheless, it is adequate to demonstrate intense-field-induced departure from ordinary behavior in the framework of nonrelativistic equations of motion, even if fully relativistic treatment would be required to obtain quantitatively reliable predictions. A further limitation on the conclusions arrived at below is that they apply to two-body systems. When a two-body system of nonzero net electric charge is immersed in an intense field, the c.m. of the system can undergo oscillations of substantial amplitude. Unless the system is in a very dilute gas, collisions occur which will negate the assumption of two-body behavior.

Some of the results developed here were prefigured in earlier papers,¹ hereafter referred to as I. Among other things, it was shown in I that the two-body Schrödinger equation in a Göppert-Mayer-type gauge (called the electric-field, or EF, gauge) becomes nonseparable in intense fields in c.m. and relative coordinates in the long-wavelength approximation (LWA). By contrast, the equations of motion are separable in Coulomb gauge² (*C* gauge) in the LWA. (The EF gauge is a simple extension of the Göppert-Mayer gauge—which is electric dipole in

nature—to a full description of the fields.) This is a result which is physically puzzling, in that the nonseparability in EF gauge could be interpreted to mean that the intense fields cause an intimate coupling of c.m. and relative motions, which should then be expected to appear in any gauge. This puzzle is shown here to arise from the use of the LWA in I when separability was examined. When a more complete description of the field is employed, the *C* gauge equations of motion also become nonseparable. The distinction between present results and those in I highlight another conclusion that was already implicit in I, that whereas LWA and electric dipole approximation are equivalent in *C* gauge, the LWA in EF gauge includes terms that go beyond electric dipole. This conclusion contradicts the commonly made presumption that LWA and electric dipole approximation are interchangeable concepts.

Section II is devoted to a demonstration that LWA and electric dipole approximation are separate and distinct concepts. This is done by first introducing EF gauge, which is a simple extension of Göppert-Mayer gauge, to a full statement of the potentials. The equations of motion in c.m. and relative coordinates are reproduced from I to show not only the loss of separability in intense fields, but also to show the presence of electric quadrupole and magnetic dipole terms even when LWA is imposed.

Section III is concerned with the failure of the usual multipole expansion when field intensity is high. This failure occurs most transparently when the net charge of the two-particle system is nonzero, but it occurs also when the total charge vanishes. Multipole expansions are examined in both EF and *C* gauges.

Some of the results from Sec. III are employed in Sec. IV to show that equations of motion in c.m. and relative coordinates become nonseparable at high field intensity. This was shown in I for EF gauge, but in *C* gauge a demonstration is done here along similar lines. By extension, such lack of separability is true in all gauges. This

loss of separability at high intensity is interpreted as an inherent field-induced coupling between relative and c.m. motions. The investigation has to be done differently when total two-body charges add up to nonzero or zero net charge.

Section V discusses briefly the phenomenon of radiation pressure, which also gives rise to a coupling between c.m. and relative coordinate motion. However, the force due to radiation pressure is a relatively weak force which gives observable consequences only over a very large number of wave periods. The intense-field effects treated here are a direct consequence of the full Lorentz forces whose effects are manifest within a single period of the plane-wave field.

II. LONG-WAVELENGTH APPROXIMATION VERSUS ELECTRIC DIPOLE APPROXIMATION

A. C Gauge

C gauge is such that a monochromatic electromagnetic plane wave is described by a zero scalar potential and a vector potential which depends only on $\omega t - \vec{k} \cdot \vec{r}$, where ω is the circular frequency of the field and \vec{k} is the field propagation vector. A simple example is

$$\begin{aligned} A_C^0 &= 0, \\ \vec{A}_C &= \vec{a} \cos(\omega t - \vec{k} \cdot \vec{r}) \end{aligned} \quad (1)$$

for a linearly polarized wave.

B. EF gauge

The gauge transformation generated by the function

$$\Lambda = \vec{A}_C \cdot \vec{r},$$

$$\begin{aligned} i\partial_t \psi(\vec{R}, \vec{r}) &= \{ -e_t \vec{R} \cdot \vec{E}(t) - e_r \vec{r} \cdot \vec{E}(t) + (1/2m_t) [-i \vec{\nabla}_R + e_t (\vec{k}/\omega) \vec{R} \cdot \vec{E}(t) + e_r (\vec{k}/\omega) \vec{r} \cdot \vec{E}(t)]^2 \\ &\quad + (1/2m_r) [-i \vec{\nabla}_r + e_r (\vec{k}/\omega) \vec{R} \cdot \vec{E}(t) + e_e (\vec{k}/\omega) \vec{r} \cdot \vec{E}(t)]^2 + V(r) \} \psi(\vec{R}, \vec{r}), \end{aligned} \quad (4)$$

where $V(r)$ represents a presumed central potential between the two particles. With subscripts 1 and 2 assigned to the two bodies, where

$$\begin{aligned} \vec{R} &= (m_1 \vec{r}_1 + m_2 \vec{r}_2) / m_t, \quad \vec{r} = \vec{r}_1 - \vec{r}_2, \\ \vec{r}_1 &= \vec{R} + m_2 \vec{r} / m_t, \quad \vec{r}_2 = \vec{R} - m_1 \vec{r} / m_t, \end{aligned} \quad (5)$$

then the masses and charges appearing in Eq. (4) are defined by

$$\begin{aligned} m_t &= m_1 + m_2, \quad m_r = m_1 m_2 / m_t, \\ e_t &= e_1 + e_2, \quad e_r = (e_1 m_2 - e_2 m_1) / m_t, \\ e_e &= (e_1 m_2^2 + e_2 m_1^2) / m_t^2. \end{aligned} \quad (6)$$

It was shown in I that the \vec{r} -dependent term in Eq. (4) which appears in association with $\vec{\nabla}_R$, and the \vec{R} -dependent term which appears with $\vec{\nabla}_r$, are both important at high field intensity. They serve to block the separation of variables in Eq. (4). Attention will now be centered on two different terms in Eq. (4), namely the R-

with \vec{A}_C as given in Eq. (1), leads to the EF gauge potentials

$$\begin{aligned} A_{EF}^0 &= -\vec{r} \cdot \vec{E}(r, t), \\ \vec{A}_{EF} &= -\frac{\vec{k}}{\omega} \vec{r} \cdot \vec{E}(\vec{r}, t), \end{aligned} \quad (2)$$

where $\vec{E}(\vec{r}, t)$ is the electric field. Equation (2) is a simple generalization (albeit not unique) of the familiar Göppert-Mayer gauge, in which the potentials consist entirely of the scalar component of Eq. (2) rendered in LWA, i.e.,

$$\begin{aligned} A_{GM}^0 &= -\vec{r} \cdot \vec{E}(t), \\ \vec{A}_{GM} &= \vec{0}. \end{aligned} \quad (3)$$

The notation $\vec{E}(t)$ in Eq. (3) means that the \vec{r} dependence in the phase $\omega t - \vec{k} \cdot \vec{r}$ has been neglected. The potentials in Eq. (3) obviously cannot be a full representation of the fields, since a time-dependent field cannot be expressed by a single-component potential. Nevertheless, Eq. (3) serves very well at long wavelengths for many purposes, unless the field is very intense. In the LWA, the \vec{E} vectors in Eq. (2) become $\vec{E}(t)$, but the vector potential remains nonzero and equivalent in importance to the scalar potential when field intensity is high.¹

C. LWA equations of motion

It was shown in I that when the two-body Schrödinger equation is written in terms of c.m. coordinates \vec{R} and relative coordinates \vec{r} , the result in EF gauge in LWA is

dependent term in the same bracket with $\vec{\nabla}_R$, and the \vec{r} -dependent vector potential term with $\vec{\nabla}_r$. When the indicated squares in Eq. (4) are carried out, both cross terms between the momentum operators and the vector potentials have vector character given by $(\vec{r} \cdot \vec{E})(\vec{k} \cdot \vec{p}_r)$, where \vec{p}_r is just the $-i \vec{\nabla}_r$ operator. The same sort of term occurs with \vec{R} and \vec{p}_R . This expression can be rewritten as³

$$\begin{aligned} (\vec{r} \cdot \vec{E})(\vec{k} \cdot \vec{p}_r) &= \frac{1}{2} [(\vec{k} \cdot \vec{p}_r)(\vec{r} \cdot \vec{E}) + (\vec{E} \cdot \vec{p}_r)(\vec{k} \cdot \vec{r})] \\ &\quad + \frac{1}{2} (\vec{E} \times \vec{k}) \cdot (\vec{r} \times \vec{p}_r), \end{aligned} \quad (7)$$

which shows that terms of this type are a combination of electric quadrupole and magnetic dipole contributions. (Note that $\vec{r} \times \vec{p}_r$ is the angular momentum operator, and for a plane wave the magnetic induction \vec{B} is $\vec{B} = \vec{k} \times \vec{E} / \omega$.) The \vec{E} in Eq. (7) is actually just $\vec{E}(t)$, reflecting the fact that Eq. (4) is an LWA expression. In other words, the leading LWA terms in ER gauge include electric quadrupole and magnetic dipole terms in addition

to the usual electric dipole contribution. Actually, the terms mixed in \vec{R} and \vec{r} dependence represent further terms beyond the electric dipole that are difficult to identify. The essential conclusion is that LWA and electric dipole approximation are different concepts. They are quite unlike each other in EF gauge. Yet the difference is gauge dependent, since LWA and electric dipole approximation amount to the same thing in C gauge.

III. FAILURE OF MULTIPOLE EXPANSIONS

A. The multipole expansion

The electric fields experienced by particles 1 and 2 in a two-body system follow from Eqs. (1) and (5) as

$$\begin{aligned}\vec{E}(\vec{r}_1, t) &= \vec{a}\omega \sin(\omega t - \vec{k}\cdot\vec{R} - m_2\vec{k}\cdot\vec{r}/m_t) \\ \vec{E}(\vec{r}_2, t) &= \vec{a}\omega \sin(\omega t - \vec{k}\cdot\vec{R} + m_1\vec{k}\cdot\vec{r}/m_t).\end{aligned}\quad (8)$$

Results for only the first element of Eq. (8) need be discussed, since the second part of Eq. (8) follows simply from the replacement $m_2 \rightarrow -m_1$. Equation (8) can be expressed as

$$\begin{aligned}\vec{E}(\vec{r}_1, t) &= \vec{a}\omega \cos(\vec{k}\cdot\vec{R} + m_2\vec{k}\cdot\vec{r}/m_t) \sin(\omega t) \\ &\quad - \vec{a}\omega \sin(\vec{k}\cdot\vec{R} + m_2\vec{k}\cdot\vec{r}/m_t) \cos(\omega t),\end{aligned}\quad (9)$$

which can be used as the starting point for a multipole expansion. A multipole expansion of Eq. (9) would normally be predicated on the hypothesis that $|\vec{k}\cdot\vec{R}| \ll 1$ and $|\vec{k}\cdot\vec{r}| \ll 1$. It will be presumed that the particles 1 and 2 are bound to each other, and that the electromagnetic field has a large wavelength as compared to a characteristic radius a_0 of the bound system. It follows that

$$|\vec{k}\cdot\vec{r}| \approx \omega a_0 \ll 1, \quad (10)$$

which can be regarded as a statement of the LWA. In I, the LWA of Eq. (10) was applied also to $\vec{k}\cdot\vec{R}$. However, the oscillations of the c.m. of a system with nonzero net

charge in an intense plane-wave field can be significant. Whereas a uniform motion of the c.m. is readily transformed away, and is of no consequence here, an oscillatory motion is important. From the known classical solutions for a charged particle in a plane wave,^{4,5} the amplitude of oscillation in the direction of the propagation vector is such that (see the Appendix)

$$|\vec{k}\cdot\vec{R}| \leq \frac{1}{4}, \quad (11)$$

with the equality in Eq. (11) approached at high field intensity. This is too large a value to permit the usual expansion hypothesis, so the leading terms in Eq. (9) are, in view of Eqs. (10) and (11),

$$\begin{aligned}\vec{E}(\vec{r}_1, t) &\approx \vec{a}\omega \cos(\vec{k}\cdot\vec{R}) \sin(\omega t) \\ &\quad - \vec{a}\omega \sin(\vec{k}\cdot\vec{R}) \cos(\omega t) \\ &\quad - \vec{a}\omega (m_2\vec{k}\cdot\vec{r}/m_t) \sin(\vec{k}\cdot\vec{R}) \sin(\omega t) \\ &\quad - \vec{a}\omega (m_2\vec{k}\cdot\vec{r}/m_t) \cos(\vec{k}\cdot\vec{R}) \cos(\omega t).\end{aligned}\quad (12)$$

Somewhat similar conclusions about coupling between relative and c.m. motions follow also from consideration of radiation reaction, which is neglected here. However, radiation pressure can be shown to be of minor consequence as compared to direct Lorentz forces, which account for the intense-field effects described here. Radiation reaction forces vis-à-vis intense-field Lorentz forces were appraised by Sarachik and Schappert.⁶ They concluded that the Lorentz forces are dominant whenever $r_0/\lambda \ll 1$ for $z < 1$, or $r_0z/\lambda \ll 1$ for $z > 1$; where r_0 is the classical electron radius, λ is field wavelength, and z is the field intensity parameter as given in the Appendix or in Eq. (19) below. These conditions are well satisfied in atomic and molecular physics problems even at extremely high field intensity.

B. EF gauge

The electromagnetic contribution to the potential energy in the Schrödinger equation in EF gauge is

$$\begin{aligned}-e_1\vec{r}_1\cdot\vec{E}(\vec{r}_1, t) - e_2\vec{r}_2\cdot\vec{E}(\vec{r}_2, t) &\approx -e_t\omega(\vec{a}\cdot\vec{R})[\cos(\vec{k}\cdot\vec{R})\sin(\omega t) - \sin(\vec{k}\cdot\vec{R})\cos(\omega t)] \\ &\quad - e_r\omega(\vec{a}\cdot\vec{r})[\cos(\vec{k}\cdot\vec{R})\sin(\omega t) - \sin(\vec{k}\cdot\vec{R})\cos(\omega t)] \\ &\quad + e_r\omega(\vec{a}\cdot\vec{R})(\vec{k}\cdot\vec{r})[\sin(\vec{k}\cdot\vec{R})\sin(\omega t) + \cos(\vec{k}\cdot\vec{R})\cos(\omega t)],\end{aligned}\quad (13)$$

to first order in $\vec{k}\cdot\vec{r}$, where Eqs. (5) and (12) have been used, in addition to the analog of Eq. (12) which relates to \vec{r}_2 .

The usual lowest-order c.m. multipole term is the electric dipole term $-e_t\omega(\vec{a}\cdot\vec{R})\sin(\omega t)$, or, equivalently, $-e_r\vec{R}\cdot\vec{E}(t)$. This is contained in the first term on the right-hand side in Eq. (13) if $|\vec{k}\cdot\vec{R}| \ll 1$. However, if the field is intense, then $\vec{k}\cdot\vec{R}$ is not small and the first two terms on the right-hand side in Eq. (13) are of similar importance. This in itself is adequate demonstration of the failure of the usual multipole expansion.

The usual lowest-order relative-coordinate multipole term arises from the third term on the right-hand side in Eq. (13). When $|\vec{k}\cdot\vec{R}| \ll 1$, this term is just $-e_r\vec{r}\cdot\vec{E}(t)$. Here again, a value of $\vec{k}\cdot\vec{R}$ which is not small destroys this identification of the leading term. When $\vec{k}\cdot\vec{R}$ is constrained only by Eq. (11), then a multipole expansion loses its meaning entirely. There is, furthermore, an intimate mixing of r and R coordinates which will be explored in the following section.

The statements made above refer to the case where $e_t \neq 0$, i.e., where the bound system exhibits a net electrical

charge. If the system is neutral, then the first two terms in Eq. (13) vanish, the magnitude of $\vec{k} \cdot \vec{R}$ is small, and the last four terms in Eq. (13) reduce simply to the simple relative-coordinate electric dipole term. It remains, how-

ever, to examine the vector potential terms in EF gauge.

The kinetic energy terms in the Schrödinger equation in EF gauge are

$$\begin{aligned}
 & (1/2m_1)[-i\vec{\nabla}_1 - e_1\vec{A}_{EF}(\vec{r}_1, t)]^2 + (1/2m_2)[-i\vec{\nabla}_2 - e_2\vec{A}_{EF}(\vec{r}_2, t)]^2 \\
 & \approx (1/2m_t)\{-i\vec{\nabla}_R + e_r\vec{k}(\vec{a} \cdot \vec{R})[\cos(\vec{k} \cdot \vec{R})\sin(\omega t) - \sin(\vec{k} \cdot \vec{R})\cos(\omega t)] + e_r\vec{k}(\vec{a} \cdot \vec{r})[\cos(\vec{k} \cdot \vec{R})\sin(\omega t) - \sin(\vec{k} \cdot \vec{R})\cos(\omega t)] \\
 & \quad - e_r\vec{k}(\vec{a} \cdot \vec{R})(\vec{k} \cdot \vec{r})[\sin(\vec{k} \cdot \vec{R})\sin(\omega t) + \cos(\vec{k} \cdot \vec{R})\cos(\omega t)]\}^2 \\
 & + (1/2m_r)\{-i\vec{\nabla}_r + e_r\vec{k}(\vec{a} \cdot \vec{R})[\cos(\vec{k} \cdot \vec{R})\sin(\omega t) - \sin(\vec{k} \cdot \vec{R})\cos(\omega t)] + e_e\vec{k}(\vec{a} \cdot \vec{r})[\cos(\vec{k} \cdot \vec{R})\sin(\omega t) - \sin(\vec{k} \cdot \vec{R})\cos(\omega t)] \\
 & \quad - e_e\vec{k}(\vec{a} \cdot \vec{R})(\vec{k} \cdot \vec{r})[\sin(\vec{k} \cdot \vec{R})\sin(\omega t) + \cos(\vec{k} \cdot \vec{R})\cos(\omega t)]\}^2. \tag{14}
 \end{aligned}$$

This result, and the potential energy terms in Eq. (13), reduce exactly to Eq. (4), (i.e., to the result found in I) when the additional assumption $|\vec{k} \cdot \vec{R}| \ll 1$ is imposed. The first comment that can be made about Eq. (14) is that, in the small $|\vec{k} \cdot \vec{R}|$ limit, electric quadrupole and magnetic dipole terms persist, as pointed out in Sec. II C. Secondly, it was shown in I in the small $|\vec{k} \cdot \vec{R}|$ limit that mixed \vec{r}, \vec{R} terms exist that are as important at high field intensity as the unmixed terms. These mixed terms cannot be given any simple multipole interpretation. Thus, even in the small $|\vec{k} \cdot \vec{R}|$ case in EF gauge, a multipole expansion loses its utility at large field intensity. However, as has been pointed out above, when $e_t \neq 0$ it is not jus-

tifiable to use a small $|\vec{k} \cdot \vec{R}|$ assumption when the field intensity is high. Instead, $\vec{k} \cdot \vec{R}$ is limited only by the higher range of the limits expressed in Eq. (11). Thus all the terms written in Eqs. (13) and (14) must be retained, and the customary version of a multipole expansion of the field has no utility at all.

C. C gauge

The Coulomb gauge potentials of Eq. (1), when referred to the coordinates of particles 1 and 2 of a two-body system, can be expanded in a fashion akin to the electric field expansion in Eq. (12). The kinetic energy terms in the two-body Schrödinger equation then become

$$\begin{aligned}
 & (1/2m_1)[-i\vec{\nabla}_1 - e_1\vec{A}_C(\vec{r}_1, \vec{t})]^2 + (1/2m_2)[-i\vec{\nabla}_2 - e_2\vec{A}_C(\vec{r}_2, \vec{t})]^2 \\
 & \approx (1/2m_t)\{-i\vec{\nabla}_R - e_r\vec{a}[\cos(\vec{k} \cdot \vec{R})\cos(\omega t) + \sin(\vec{k} \cdot \vec{R})\sin(\omega t)] + e_r\vec{a}(\vec{k} \cdot \vec{r})[\sin(\vec{k} \cdot \vec{R})\cos(\omega t) - \cos(\vec{k} \cdot \vec{R})\sin(\omega t)]\}^2 \\
 & + (1/2m_r)\{-i\vec{\nabla}_r - e_e\vec{a}[\cos(\vec{k} \cdot \vec{R})\cos(\omega t) + \sin(\vec{k} \cdot \vec{R})\sin(\omega t)] + e_e\vec{a}(\vec{k} \cdot \vec{r})[\sin(\vec{k} \cdot \vec{R})\cos(\omega t) - \cos(\vec{k} \cdot \vec{R})\sin(\omega t)]\}^2. \tag{15}
 \end{aligned}$$

The lowest-order terms when $|\vec{k} \cdot \vec{R}| \ll 1$ are just the LWA or electric dipole approximation terms. These are the same in this gauge, unlike the EF gauge. These leading terms follow from $\vec{a}\cos(\vec{k} \cdot \vec{R})\cos(\omega t)$ in Eq. (15), where that term has the coefficient e_t in the curly bracket with $-i\vec{\nabla}_R$ and the coefficient e_r in the bracket with $-i\vec{\nabla}_r$. When $|\vec{k} \cdot \vec{R}|$ is not small, then $e_t\vec{a}\sin(\vec{k} \cdot \vec{R})\sin(\omega t)$ and $e_r\vec{a}\sin(\vec{k} \cdot \vec{R})\sin(\omega t)$ are of similar importance to the electric dipole term. Therefore, these extra terms destroy the usual multipole expansion, even without reference to the terms proportional to $\vec{k} \cdot \vec{r}$ in Eq. (15).

The above conclusion holds true when $e_t \neq 0$. When $e_t = 0$, not only does that portion of Eq. (15) proportional to e_t vanish, but then $|\vec{k} \cdot \vec{R}| \ll 1$ because there is no net charge on the bound system. In this case, however, the curly bracket with $-i\vec{\nabla}_R$ contains

$$-e_r\vec{a}(\vec{k} \cdot \vec{r})\cos(\vec{k} \cdot \vec{R})\sin(\omega t) \approx -e_r(\vec{k} \cdot \vec{r}/\omega)\vec{E}(t). \tag{16}$$

This leads to an interaction term in the Hamiltonian given by

$$H_1 = -e_r(\vec{k} \cdot \vec{r}/\omega)[\vec{p}_R \cdot \vec{E}(t)]/m_t, \tag{17}$$

which is to be compared to the $P_R^2/2m_t$ arising from the same bracket in Eq. (15) containing the $-i\vec{\nabla}_R$ (or \vec{p}_R) operator. A general order-of-magnitude comparison of these two contributions leads to

$$\frac{|e_r(\vec{k} \cdot \vec{r}/\omega)[\vec{p}_R \cdot \vec{E}(t)]/m_t|}{P_R^2/2m_t} = O\left(\frac{2e|\vec{E}|a_0}{|\vec{p}_R|}\right), \tag{18}$$

where a_0 is a characteristic size of the bound system. The result (18) is precisely of the form of Eq. (68) of I, and so the conclusions reached in I hold here as well. Specifically, it was shown in I that Eq. (18) is proportional to $z^{1/2}$, with a factor of proportionality close to unity, where z is the intensity parameter⁷

$$z = e^2 a^2 / 2m^2, \quad (19)$$

and a is the amplitude of \vec{a} . Thus the interaction term in Eq. (17) is of major importance in an intense field. Since this term bears no relationship at all to a conventional multipole expansion of the field, the final conclusion is that high field intensity causes the usual multipole expansion to lose all utility whether $e_t \neq 0$ or not.

IV. FAILURE OF SEPARABILITY

A. EF gauge

The information necessary to assess separability of the c.m. and relative-coordinate equations of motion has already been established. Although mixed \vec{r}, \vec{R} terms exist in Eqs. (13) and (14) beyond those that were identified in I, nevertheless the separation-blocking terms identified and appraised in I are sufficient to establish the essential conclusion. This conclusion is that at high field intensity [when z as given in Eq. (19) is within 2 or 3 orders of magnitude of unity] the equations of motion in EF gauge, expressed in c.m. and relative coordinates, cannot be separated. Physically, this is because the c.m. and relative motions become coupled when the field is intense.

B. C gauge

In I only LWA terms were retained, and the equations of motion were found to be separable in C gauge. Terms beyond the LWA are presented in Eq. (15). In the expression in Eq. (15) that contains the $-i\vec{\nabla}_r$ operator, it is the term $-e_r \vec{a} \cos(\vec{k} \cdot \vec{R}) \cos(\omega t)$ which is responsible for the LWA or electric dipole approximation term when $|\vec{k} \cdot \vec{R}| \ll 1$. When $e_t \neq 0$ and the field intensity is high enough that $|\vec{k} \cdot \vec{R}|$ is in the upper part of the range in Eq. (11), then the term $-e_r \vec{a} \cos(\vec{k} \cdot \vec{R}) \cos(\omega t)$ acquires some \vec{R} dependence. Even more important is the fact that $-e_r \vec{a} \sin(\vec{k} \cdot \vec{R}) \sin(\omega t)$ achieves as much significance as the LWA term and has strong \vec{R} dependence. Since this \vec{R} -dependent term appears in direct association with the $-i\vec{\nabla}_r$ operator, Eq. (15) is clearly nonseparable.

When $e_t = 0$, then one must consider $|\vec{k} \cdot \vec{R}|$ to be small. Attention will now be directed to the expression in Eq. (15) that contains $-i\vec{\nabla}_R$. With the hypotheses $e_t = 0$ and $|\vec{k} \cdot \vec{R}| \ll 1$, the expression in question reduces to

$$[-i\vec{\nabla}_R - e_r \vec{a}(\vec{k} \cdot \vec{r}) \sin(\omega t)]^2 = [-i\vec{\nabla}_R - e_r(\vec{k} \cdot \vec{r}/\omega)\vec{E}(t)]^2. \quad (20)$$

The issue of the significance of the field-dependent term in Eq. (20) was discussed in Sec. III C above [see Eq. (16)], where it was concluded that high field intensity caused this term to be very important. This is then another example of a mixed \vec{R}, \vec{r} term which blocks separation of variables. That is, high field intensity causes the two-body equations of motion in C gauge to become nonseparable whether $e_t \neq 0$ or not.

V. COMPARISON WITH RADIATION PRESSURE EFFECTS

It is known (see, for example, Refs. 8 or 9) that radiation pressure will also induce a coupling between c.m. and relative coordinates. However, radiation pressure effects and the explicit intensity effects considered here operate through different mechanisms and on different time scales.

Radiation pressure arises from the fact that momentum transfer between the atom and a plane-wave electromagnetic field is collinear with the field propagation direction for photon absorption and for induced emission; but for spontaneous emission the recoil is random. This leads to a net force on the atom in the \vec{k} or $-\vec{k}$ direction, depending on the relative populations of ground and excited states. To set an order of magnitude, suppose the radiation pressure force on a two-level atom has reached its strong-field saturation value of $A\vec{k}/2$,⁹ where A is the Einstein A coefficient. This force will produce an acceleration of the atom which, in the course of a single wave period, produces a displacement R given by

$$kR = A\pi^2/m_t. \quad (21)$$

For order-of-magnitude purposes, one can set $A \approx 10^8 \text{ sec}^{-1}$ and replace m_t by the mass of a hydrogen atom. The result is

$$kR \approx 10^{-15}. \quad (22)$$

As Eqs. (21) and (22) show, radiation pressure gives rise to quite small forces which make their effects felt only after very many wave periods. On the other hand, the intense-field effects treated here arise directly from the full Lorentz forces, and they have major consequences within a single period of the electromagnetic field. For instance, the limit expressed in Eq. (11) represents an oscillation of the atom's c.m. in a single wave period in the frame in which the c.m. is, on average, at rest. It may thus be compared directly with Eq. (22), which represents the c.m. motion in one wave period, starting from rest. Eventually, of course, the acceleration due to radiation pressure can have macroscopic consequences, but the disparity between Eqs. (11) and (22) means that intense-field effects and radiation-pressure effects are properly viewed as independent phenomena.

There is a physical picture one can form of the intense-field coupling of c.m. and relative motions which follows from the figure-eight behavior reviewed in the Appendix. This figure-eight motion constitutes an intrinsic angular momentum transferred to the charged particle by the field. This is in contrast to the linear, zero-angular-momentum motion executed (in the frame where the particle is at rest on average) by a charged particle in a quasi-static electric field or in a low-intensity plane wave. The intrinsic angular momentum of the particle motion in an intense field can be thought of as leading directly to higher-order multipole moments in the motions of the charged particles constituting the bound system, thus giving rise directly to c.m. and relative-coordinate coupling.

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APPENDIX

An exact classical solution for the motion of a free, charged particle in a linearly polarized plane-wave electromagnetic field is given in Refs. 4 and 5. In the frame of reference in which the particle is, on the average, at rest, the particle follows a figure-eight trajectory. With the y axis along the direction of polarization, and the x axis along the direction of propagation of the field, the solution is

$$\omega y = -\xi^{1/2} \cos(\omega t - \omega x), \quad (\text{A1})$$

$$\omega x = -\frac{1}{8} \xi \sin 2(\omega t - \omega x), \quad (\text{A2})$$

where

$$\xi \equiv 2z/(1+z). \quad (\text{A3})$$

The electric field vector and magnetic induction vector that give rise to Eqs. (A1) and (A2) are

$$\vec{E} = \vec{a} \omega \cos(\omega t - \vec{k} \cdot \vec{r}),$$

$$\vec{B} = (\vec{k}/\omega) \times \vec{E}.$$

The quantity z in Eq. (A3) is the same intensity parameter defined in Eq. (19).

The amplitude of the oscillatory motion in the \vec{k} direction is, from Eqs. (A2) and (A3),

$$|\omega x|_{\max} = z/4(1+z). \quad (\text{A4})$$

For high field intensity, Eq. (A4) yields

$$\lim_{z \rightarrow \infty} |\omega x|_{\max} = \frac{1}{4}. \quad (\text{A5})$$

Since the x axis is aligned with \vec{k} , Eq. (A5) gives rise to the result in Eq. (11),

$$|\vec{k} \cdot \vec{R}| \leq \frac{1}{4}.$$

¹H. R. Reiss, Phys. Rev. A **19**, 1140 (1979); **22**, 770 (1980).

² C gauge was referred to as R gauge (radiation gauge) in I.

³Note that $(\vec{r} \cdot \vec{E})(\vec{k} \cdot \vec{p}_r) = (\vec{k} \cdot \vec{p}_r)(\vec{r} \cdot \vec{E})$ because $\vec{k} \cdot \vec{E} = 0$ for the transverse plane-wave fields considered here.

⁴E. S. Sarachik and G. T. Schappert, Phys. Rev. D **1**, 2738 (1970), Eq. (2.29).

⁵L. D. Landau and E. M. Lifshitz, *The Classical Theory of Fields* (Pergamon, Oxford, 1975), p. 118, solution to problem 2.

⁶E. S. Sarachik and G. T. Schappert, Phys. Rev. D **1**, 2738 (1970), Eqs. (3.60a) and (3.60b).

⁷This quantity was labeled z_m in I, where it was defined in more general terms than in Eq. (19). As stated, z depends on the C gauge vector potential. A gauge-independent general definition of z is $z = e^2 \langle \vec{E}^2 \rangle / m^2 \omega^2 = e^2 \langle \vec{B}^2 \rangle / m^2 \omega^2$, where the angular brackets indicate a time average over a wave period.

⁸V. S. Letokhov and V. G. Minogin, Phys. Rep. **73**, 1 (1981).

⁹R. J. Cook, Comments At. Mol. Phys. **10**, 267 (1981).