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Single-Cycle Electron Acceleration in Focused Laser Fields*

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The possibility of accelerating particles with a focused laser is considered. The particle is accelerated *transversely* by the electric field and redirected to the forward direction within a single half-cycle by the magnetic field of the radiation. The equations of motion of the particle are solved and discussed. Special attention is given to an intuitive understanding of the constraint imposed by the geometry of the focal region. Optimum conditions are examined.

I. INTRODUCTION

One of the most basic problems of the interaction of radiation and matter is the motion of a charged particle in a plane electromagnetic wave. We shall solve this problem classically, and apply the results to the acceleration of electrons to highly relativistic energies using the intense fields of pico-second laser pulses. We will approximate the high-intensity region of a focused beam by a plane wave, and discuss the problems and the errors this introduces. This procedure enables valuable insight into the nature of the problem, and gives numerical results to within a factor of 2. The order of magnitude of laser power readily available is 10^{12} W, which means that a diffraction-limited focus (take $\lambda = 1 \mu$) gives fields of the order of 10^9 esu or 3×10^{11} V/cm. In a single pass across such a focus, an electron could gain in energy 3×10^7 eV. Since this implies that the electron becomes relativistic within a small fraction of an optical cycle, the usual linear approximation, which ignores the magnetic field of the wave and results in simple sinusoidal oscillation of a particle, is no longer valid. Once the electric field of the plane wave has given the particle a large transverse velocity, the magnetic field causes the particle's path to be bent into the direction of travel of the wave. In very large fields, the particle's velocity then approaches c in the forward direction very quickly and it tends to travel with the wave, gaining energy from it. When the particle has finally fallen behind the wave enough so that the field directions are reversed, it is symmetrically decelerated.

This acceleration mechanism is currently of great astrophysical interest because of the enormous energies that can be transferred to particles.¹ This idea was suggested years ago by Menzel and

Salisbury² and by McMillan³ for the origin of cosmic rays, with the solar corona as the source of the electromagnetic waves. Gunn and Ostriker⁴ recently conjectured that cosmic rays are generated by pulsars by this same mechanism. Proton energies of up to 3×10^{17} eV can be expected from a typical pulsar. Gunn and Ostriker estimate the magnetic dipole radiation fields caused by the rotation of neutron stars to be on the order of 10^{10} esu, which is only an order of magnitude larger than obtainable in the focused laser. The wavelength of the radiation for the Crab pulsar, however, is 10^9 cm, which is a factor of 10^{13} larger than laser wavelengths. Since Gunn and Ostriker's calculations indicate that the energy of the particle increases as $m^{1/3} \lambda^{2/3} E^{2/3}$, where E is the electric field, we can expect 10^7 -eV electrons from the same mechanism with laser fields, which supports our crude estimate above. These results are especially encouraging considering that peak laser power might possibly become several orders of magnitude higher than at present; peak powers have been increasing by orders of magnitude in the past few years, and there seems to be no fundamental limit on these peak powers. Hence it seems worth examining more closely the possibilities of this particularly simple and promising laser acceleration mechanism.

II. TRAVELING AT SPEED OF LIGHT OR SOLUTION BY INTRODUCTION OF INDEX OF REFRACTION

Although the problem of the classical motion of a charged particle in a plane electromagnetic wave has been solved many times,^{5,6} it is so fundamental and interesting a problem that we think it worthwhile to report a novel method of solution. Though lengthier in simple cases, this method is more physically intuitive because it eliminates the need to solve parametrically in terms of $\eta = \omega t - kz$, as

required by other methods. Also our method of solution allows the treatment of phase velocity other than c , as in a focused beam or in an interstellar plasma.

To solve this problem we imagine a medium with an index of refraction $n > 1$. We then transform into a reference frame moving at the velocity of light in that medium, solve for the motion of the particle in that frame, transform back into the laboratory frame, and then take the limit $n \rightarrow 1$. No property of the medium other than its index of refraction is considered. In the medium the light can be treated as traveling at speed c/n so that it is actually possible to observe the interaction from a reference frame co-moving with the light wave. In such a frame we will find that the fields do not change with time; the problem is reduced to that of the motion of a charged particle in a *static* (but nonuniform) magnetic field. The limiting process is a completely physical one corresponding to the introduction and then evacuation of a thin gas from the region of interaction, so we can expect no mathematical problems with taking limits. With this method we have found the motion of the particle for arbitrary injection energy, direction, and phase with respect to the wave.

The simplest possible electromagnetic field to use in this problem is

$$\begin{pmatrix} \vec{E} \\ \vec{B} \end{pmatrix} = \begin{pmatrix} \hat{i} E \\ \hat{j} B \end{pmatrix} \cos(kz - \omega t). \quad (1)$$

The solution for a more complex waveform cannot be given by superposition in this nonlinear problem, but can be found in a manner similar to this simple case. Postulating a thin medium, with index of refraction $n \gtrsim 1$, requires

$$k = n\omega/c, \quad B = nE. \quad (2)$$

The light is traveling through this medium at a velocity c/n in the positive \hat{z} direction. We wish to travel along with the light, and therefore to transform from the laboratory frame to a reference frame moving with velocity $v_n = c/n$ in the positive \hat{z} direction. Hence let us define

$$\beta_n = 1/n, \quad \gamma_n = (1 - \beta_n^2)^{-1/2}. \quad (3)$$

We call this new frame, in which all quantities are primed, the "light frame." The quantity $kz - \omega t$ corresponds to the number of crests of the electromagnetic wave between two physical space-time points and so must be an invariant. Thus $kz - \omega t = k'z' - \omega't'$, and (\vec{k}, ω) is a four-vector. Therefore, we have

$$\omega' = \gamma_n(\omega - ck\beta_n) = \gamma_n\omega(1 - \beta_n n) = 0,$$

$$k' = \gamma_n(k - \beta_n\omega/c) = \gamma_n k(1 - \beta_n/n) = k/\gamma_n.$$

In the light frame the electromagnetic fields are

static, since the wave has been Doppler shifted to zero frequency. Spatial variation is still present, but much more gradual as $k' \ll k$. Note that the index of refraction in the light frame $n' = ck'/\omega' = \infty$.

The magnitudes of the field become

$$E' = \hat{i}\gamma_n(E - \beta_n B) = 0, \quad B' = \hat{j}\gamma_n(B - \beta_n E) = B/\gamma_n,$$

using Eqs. (2) and (3). As viewed from the new frame, then, the particle is moving in a static magnetic field $\vec{B}' = \hat{j}(B/\gamma_n) \cos k'z'$. The electric field has vanished.

In the light frame, the relativistic equation of motion for the four-velocity of the particle, $u'^\mu = (\gamma' \vec{\beta}', \gamma')$, is

$$\frac{du'^\mu}{d\tau} = \frac{e}{mc} F'^{\mu\nu} u'_\nu,$$

where τ is the proper time. This immediately gives

$$\begin{aligned} \frac{du'_x}{dt'} &= -\omega'_c u'_z \cos k'z', \\ \frac{du'_z}{dt'} &= \omega'_c u'_x \cos k'z', \end{aligned} \quad (4)$$

$$\frac{du'_y}{dt'} = \frac{d\gamma'}{dt'} = 0,$$

where $\omega'_c = eB'/\gamma'mc$. Thus the energy of the particle, $\gamma'mc^2$, is a constant of the motion in the light frame. This is of course because a purely magnetic field cannot do work. Hence γ' and β' are constants of the motion.

To integrate these equations, the initial conditions must be specified. In the simplest case the particle is initially stationary at zero phase of the wave in the laboratory; more general conditions add no conceptual complications. With these simple initial conditions, Eq. (4) integrates to

$$u'_x = -\Lambda \sin k'z', \quad (5)$$

where $\Lambda \equiv \omega'_c \gamma' / k' c = eB/mc^2 k = neE/mc^2 k$ measures the strength of the interaction. Equation (5) implies

$$u'_z = u'(1 - \epsilon \sin^2 k'z')^{1/2}, \quad (6)$$

where $\epsilon \equiv (\Lambda/u')^2$ and where $u' = \gamma'\beta'$. Note that the initial conditions require $u' = -\gamma_n \beta_n$. The particle is injected *backwards* very rapidly in the light frame, and is curved off axis by the static magnetic field, without any change in its total energy. For a very thin medium ϵ is much less than unity, going to zero in a vacuum. Therefore Eq. (6) becomes

$$\beta' c \approx \frac{dz'}{dt'} \left(1 + \frac{1}{2} \epsilon \sin^2 k'z'\right),$$

which can be integrated to give

$$z' + v_n t' \approx (\epsilon/4k') \left[\eta - \frac{1}{2}(\sin 2\eta)\right],$$

where $\eta \equiv \omega t - kz = -k'z'$, the phase distance the particle falls behind the wave in the course of the motion. Transforming back into the laboratory frame and letting $n \rightarrow 1$ gives

$$kz = \frac{1}{4}\Lambda^2[\eta - \frac{1}{2}(\sin 2\eta)] . \quad (7)$$

Likewise, Eq. (5) implies

$$dx' = -\Lambda \sin(k'z') dz' / \gamma' \beta'_z .$$

Ignoring terms of order ϵ , this can be integrated to give, in the laboratory frame,

$$kx = \Lambda(1 - \cos \eta) . \quad (8)$$

We note that $u_x = \gamma_n(u'_x + \beta_n \gamma' v'_y)$, expressing u'_x in terms of u'_y , immediately implies the useful relation

$$u_x = \frac{1}{2}u_x^2 . \quad (9)$$

Since $\gamma^2 = 1 + u^2$, we can use Eqs. (9) and (5), and the fact that $u_x = u'_x$ is the transverse component of a four-vector, to find

$$\gamma = 1 + u_x = 1 + \frac{1}{2}\Lambda^2 \sin^2 \eta . \quad (10)$$

Thus the particle gains energy as η increases, that is, as it falls behind the wave, until η reaches $\frac{1}{2}\pi$. The energy gained and the forward distance traveled are independent of the sign of the charge of the particle, as is intuitively reasonable. In the light frame the curl of \vec{B}' is clearly nonzero and yet the electric field is zero. This is maintained by the polarization currents present in the medium. Such considerations are implicit in a simple transformation of fields.

III. CONSTANTS OF MOTION

A straightforward solution of the motion of a charged particle in a plane electromagnetic wave is achieved by examining the constants of the motion of the particle. This method is essentially the Hamilton-Jacobi solution of Landau and Lifshitz⁵ put into a more explicit form. The equations of motion for a particle in the fields of Eq. (1) in vacuum are

$$\frac{d(p_x)}{d\tau} = \frac{eE \cos \eta}{mc} \left(\frac{W}{c} - p_x \right) , \quad (11)$$

$$\frac{d(p_y)}{d\tau} = \frac{eE \cos \eta}{mc} (0) , \quad (12)$$

$$\frac{d(p_z)}{d\tau} = \frac{eE \cos \eta}{mc} (p_x) , \quad (13)$$

$$\frac{d(W/c)}{d\tau} = \frac{eE \cos \eta}{mc} (p_x) . \quad (14)$$

By inspection and minor manipulation of these equations we can find the constants of the motion

$$W - cp_x = mc^2 \delta , \quad (15)$$

$$\frac{1}{2}p_x^2 - mc\delta p_x = K , \quad (16)$$

$$p_y = K' , \quad (17)$$

$$p_x + (e/c)A_x = K'' , \quad (18)$$

where δ , K , K' , and K'' are constants defined by the initial conditions, and A_x is the vector potential, which for these fields is given by

$$A_x = -(cE/\omega) \sin \eta . \quad (19)$$

Equations (15) and (16) are not really independent as they are related by the condition that

$$W^2 - p_x^2 c^2 - p_y^2 c^2 - p_z^2 c^2 = m^2 c^4 .$$

These equations for the constants of the motion specify the problem completely. With the simple initial conditions used above, Eq. (18) immediately becomes

$$p_x = \Lambda mc \sin \eta . \quad (20)$$

Combining this with Eqs. (15) and (16) gives an energy gain corresponding to Eq. (10), which can be integrated parametrically in terms of η to give Eq. (7). These results then are the same as those found by introducing a refractive medium.

The methods described above can be easily extended to more generalized initial conditions. The particle begins at the origin but at phase ϕ of the wave, rather than at zero phase where the fields are strongest. It is injected with energy $\gamma_i mc^2$, with velocity $\beta_i c$ at the angle from the forward direction whose tangent is $\alpha \equiv \beta_{ix}/\beta_{iz}$. Equations (10) and (7) then generalize to

$$\begin{aligned} \Delta\gamma &= \gamma - \gamma_i \\ &= (\Lambda \beta_{ix} \gamma_i / \delta) [\sin(\eta - \phi) + \sin \phi] \\ &\quad + (\Lambda^2 / 2\delta) [\sin(\eta - \phi) + \sin \phi]^2 , \end{aligned} \quad (21)$$

$$\begin{aligned} kz &= \gamma_i \beta_{iz} \eta / \delta + (\Lambda \gamma_i \beta_{ix} / \delta^2) \\ &\quad \times [(1 - \cos \eta) \cos \phi + (\eta - \sin \eta) \sin \phi] \\ &\quad + (\Lambda^2 / 8\delta^2) [- (2\eta + \sin 2\eta - 4 \sin \eta) \cos 2\phi \\ &\quad + (3 + \cos 2\eta - 4 \cos \eta) \sin 2\phi \\ &\quad + 4(\eta - \sin \eta)] , \end{aligned} \quad (22)$$

where $\delta = \gamma_i (1 - \beta_{iz})$, as before. It must be emphasized that this is not an explicit solution, as η depends upon z .

The constants of motion [Eqs. (15)–(18)] can be understood physically. The constant δ is the (dimensionless) momentum of the particle in the $z + ct$ direction, as can be seen from a Hamilton-Jacobi solution to the problem.⁶ The Hamiltonian of a charged particle in an electromagnetic wave traveling in the $+z$ direction is a function of the $z - ct$

coordinate only and not of the $z+ct$ coordinate; the momentum in the $z+ct$ direction must then indeed remain constant. The Minkowski force upon the particle at any instant, $dp_x/d\tau$, is equal to $eE(t)\delta$. In the particle's instantaneous rest frame it feels an acceleration proportional to the strength of the field, so that δ is a measure of the particle's susceptibility to being accelerated. For example, if the particle is injected energetically in the $+z$ direction it will have $\delta \sim 0$, and will be little affected by the fields. In a sense, the particle can only gain energy to the extent that it lags behind the wave, but in this case it stays very close to it. This is in contrast to a particle in a linear accelerator, where there is a longitudinal component of the electric field to continue the acceleration at velocities tantamount to c .

Equation (16) reflects the fact that the force in the forward direction, dp_z/dt , is contributed entirely by the magnetic field, and so is directly proportional to the sideways velocity of the particle, v_x . Equations (17) and (18) simply state that for directions in which the field does not vary, the corresponding canonical momenta are conserved. Both of these equations can be derived directly from the equations of motion using the defining equation (19).

IV. LASER ACCELERATOR

Is it practical to use intense laser fields to accelerate electrons to high energies? Equation (21) indicates that energy gains ($\Delta\gamma mc^2$) which are proportional to $\Lambda^2\gamma_i$ might be achieved, because $\delta \sim (2\gamma_i)^{-1}$ for relativistic forward injection. Since gigawatt laser pulses can give values of Λ of around 10, this would mean multiplying the energy of the incident particle by 100. But such intense fields require focusing the beam, and so can only be maintained over a short distance. Multiplicative energy gains are impossible: To accelerate particles with large initial velocities requires great distances, as Eq. (22) makes clear. We are forced to consider an optimization procedure, to balance the strong fields present at a sharp focus with the longer distances available with less focusing. For the sake of calculation we approximate the complex high-intensity region of the focused beam by a plane wave of extent $4F^2\lambda$ along the beam and $F\lambda$ across it,⁷ as in Fig. 1. F is the F number, or nominal focal ratio. The constriction of the beam is limited by diffraction. The limitations of this approximation are discussed below. Our results are exactly applicable to an unfocused plane wave, which will diverge because of diffraction.

The experimental variables are F and the power P :

$$P = (c/8\pi) E^2 (F\lambda)^2.$$

We define the dimensionless parameter

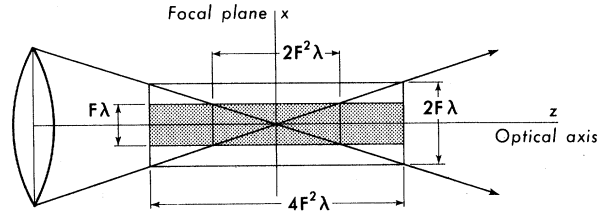


FIG. 1. Geometry of the focus of a lens with nominal focal ratio F . The intense field region scales as F^2 longitudinally and as F transversely.

$$D = 2F\Lambda = (e/mc^2)(8P/\pi c)^{1/2}. \quad (23)$$

With the moderate power of 2.5×10^{11} W, $D = 8.55$ does not vary with F and is a direct measure of the optical power. We first consider the problem on simple physical grounds. The energy gained by the electron in crossing the focus is $\Delta\mathcal{E} = \text{force} \cdot \Delta x$, so that $\Delta\gamma = eE\Delta x/mc^2$, if the fields were to remain at their strongest. But $eE/mc^2 = k\Lambda = kD/2F$. If the maximum allowable Δx is $F\lambda$, then the maximum $\Delta\gamma$ to be attained is πD . This simple analysis indicates that the energy to be gained by this acceleration technique is strictly limited, regardless of the injection velocity, and can only be increased as the square root of the available power. In this picture the higher fields obtained by focusing are only at the expense of shorter possible distances across the focus. Note that focusing does not increase the maximum energy available to the particle, πDmc^2 , which depends only upon the total power. Focusing does affect the distribution of that power, and so determines how close the actual energy gain approaches the limit. The limiting energy gain does not depend on the wavelength of the laser, for a given power. The more exact calculations verify these conclusions.

The electron is injected into the focal region with an initial energy γ_i at an angle to the forward arc α , in the plane of the electric field. We will assume that for cases of interest the electron leaves the focal region through the front rather than through the side, so that the limiting equation is $kz = 8\pi F^2$ instead of $kx = 2\pi F$. Since this acceleration mechanism results in particle paths being bent to the forward, this assumption clearly holds if $\alpha < 1/4 F$. We find below that even when $\alpha > 1/4 F$ the optimum trajectory particle leaves the focal region near the corner, without an explicit restriction in the x direction.

We wish to find the maximum energy that can be gained by the particle for a given set of initial conditions. Experimentally we would vary the F number, making a narrower or a broader focus to find the optimum geometry. But mathematically, since Eqs. (21) and (22) are such complicated functions

of η , we will have to implicitly vary F by varying η . Remembering that the particle leaves the focal regions at $kz = 8\pi F^2$, Eq. (22) [using Eq. (23)] becomes a quartic for F in terms of η . This can be solved explicitly for $F = F(\eta)$. F determines the fields because it gives the area that the laser power is spread across, and it also determines the distance the particle can travel in these fields. Therefore, for a given set of initial conditions, F fixes the motion of the particle and hence the phase lag η of the particle as it leaves the focal region. Equation (21) then becomes $\Delta\gamma = \Delta\gamma(F(\eta), \eta)$, understood to refer to the exiting values. A computer maximization of this equation with respect to η gives the largest $\Delta\gamma$ for the specified initial conditions, as well as the F number required for this optimum.

This optimization procedure was carried out to find the energy gain $\Delta\gamma mc^2$ for a wide range of initial conditions. $\Delta\gamma$, for a particle injected at the peak of the wave (phase C in Fig. 2), at the moderate power of 2.5×10^{11} W, is shown as a function of injection angle and energy in Fig. 3(a). The corresponding optimizing F number is shown in Fig. 3(b).

Only the injection phases between C and E (Fig. 2) were considered. The phase point F is equivalent to C in that a particle beginning at either of those points would experience the same forwards acceleration. If the particle is started between F and E , it will begin to be decelerated as soon as it falls behind the waves as far as E , and so cannot reach the higher-energy gains. We find that for any initial conditions, regardless of the phase at which it is started, the particle must leave the focal region at phase B in order to maximize its acceleration, where B is in a narrow range of phase between 60° and 70° behind C . Likewise the energy gain is greatest when the particle enters the beam at phase D , within a similar range ahead of C . By entering and leaving the focal region at these points, the particle stays in the beam only so long as the fields are strongest and so receives the maximum energy. This is a soft maximum, though, and the energy gained varies by less than a factor of 2, in general considerably less, when the injection phase is

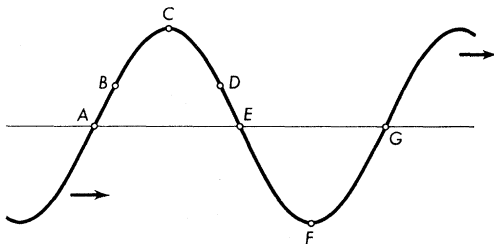


FIG. 2. Single cycle of electromagnetic radiation. The letters are referred to in the text.

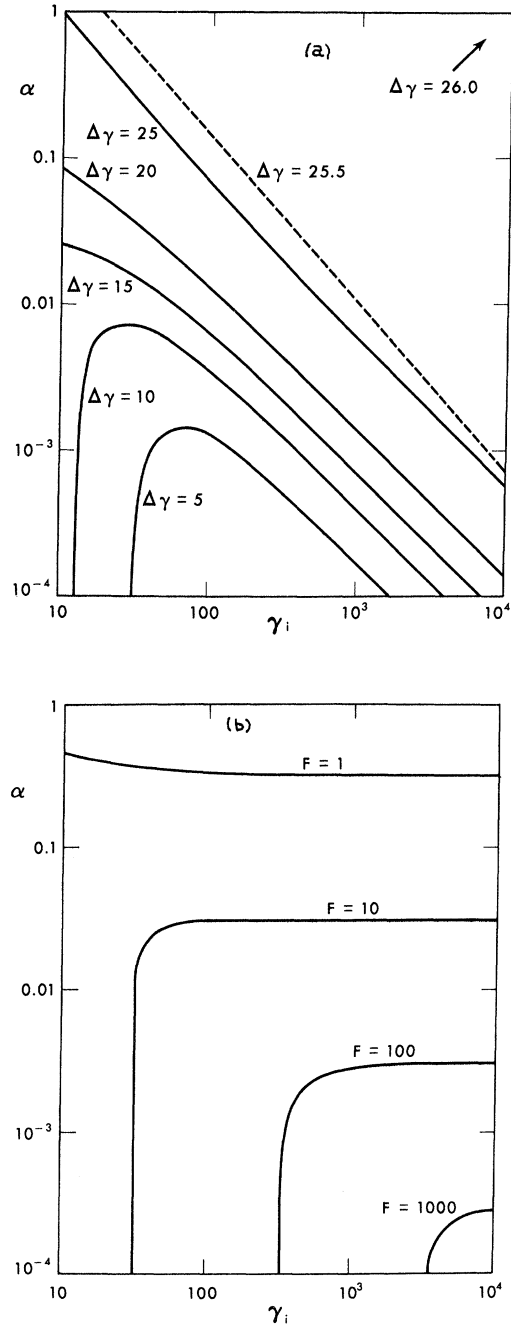


FIG. 3. Computer results of (a) optimum normalized energy gain $\Delta\gamma$ and (b) optimum F number, as a function of normalized injection energy γ_i and of the tangent of the injection angle to the forward, α . The results shown are for laser power $P = 2.5 \times 10^{11}$ W, with the particle injected at the peak of the optical cycle ($\phi = 0$).

varied between C and E .

The most striking result of the optimization is that at a given power level there is a maximum energy gain which cannot be surpassed. Figure 3(a)

indicates that for $P = 2.5 \times 10^{11}$ W an electron injected at the peak of the wave cannot possibly attain a $\Delta\gamma$ greater than 26. This result supports the simple model considered above. The magnetic field of the wave does not accelerate the particle; it can only change its direction. The electric field does the accelerating, but only *across* the focus. Thus the energy gain is strictly limited by the finite width of the focus. Figure 3(a) also indicates that it is generally advantageous to inject the particle at a slight angle to the forward. This is because, for a highly relativistic particle moving directly forward, the forces due to \vec{E} and $\vec{v} \times \vec{B}$ balance each other almost exactly, and little acceleration results. But if the particle is moving at a slight angle to the forward, there is a significant component of the magnetic force in the forward direction. In other terms, the particle injected at an angle will be able to move further across the focal region before leaving through the front. For a less relativistic particle, whose injection energy does not overwhelm its subsequent energy gain, this effect is less important, as can be seen in the shape of the curves in Fig. 3(a).

It is clear that a number of the subtleties of this problem have been ignored. The high-intensity region of a focused beam is quite definitely *not* a sharply delimited plane wave. The intensity is actually a maximum at the very focal point, falling off in every direction. Superimposed upon an actual focus, our chosen homogeneous focal region extends to about the half-intensity circle in the focal plane, and to about the 80% maximum intensity points along the optical axis. The intensity in our focal region is $4/\pi$ times the actual intensity at the focal point of a focused beam of equal power.⁷ The numerical factors in the dimensions of our focal region were of course chosen intuitively, to give a reasonable representation of the focus. The more important consideration is the scaling of the focal dimensions with F , which we express exactly. We have also totally ignored the effects of the particle's entering and leaving the interaction region, which are expected to be minimal. These simplifications should not fundamentally change the nature of the problem, but only modify the numerical results by a small factor.

A more serious problem we have ignored in using our plane-wave approximation is the speeding up of the wave through the focus. In the focal region, the k vector is decreased by a factor of $1 - 1/16F^2$ compared with a parallel beam of light of the same wavelength.⁷ This is to be expected, for when the phase velocity of a plane wave of velocity c is mea-

sured along a diagonal to the direction of travel, it is greater than c . This effect is alternatively thought of as the Gouy phenomenon: the decrease in phase by π of a ray through the focus. For our purposes, this means that the wave will more rapidly outdistance the particle, decreasing the time for effective acceleration. Since the wave does not travel at c , it might be difficult to accelerate the particle to as close to c as desirable.

Intuitively, this should not be an overwhelming effect. It is the spreading of the focused beam that causes the speeding up, just as it causes the finite length of the intense field region. We might then expect these two effects to be of similar weight in limiting acceleration. In fact, we can try to express the speeding up by decreasing the effective acceleration distance by a small numerical factor. To see this quantitatively, we find the distance z_p in which the actual focused beam surpasses our ideal plane wave in phase by $\frac{1}{2}\pi$, at which time the particle will certainly be out of phase:

$$\begin{aligned} [k - (1 - 1/16 F^2)k] z_p &= \frac{1}{2}\pi, \\ (k/16 F^2) z_p &= \frac{1}{2}\pi, \\ z_p &= 4 F^2 \lambda. \end{aligned}$$

We see that acceleration is curtailed by the speeding up about as quickly as it is curtailed by leaving the intense field region. This effect then cuts practical energy gain by something like a factor of 2.

Another way of seeing this is to notice that the speeding up of the wave is significant only when it is the main contributor to the particle's lagging the wave. This is the case only when the particle is traveling very close to c : When the particle's velocity $\beta \approx 1 - 1/16 F^2$. This condition implies $\gamma \approx 2\sqrt{2}F$. We find in our computer calculations for the idealized plane wave, in fact, that for any injection energy the optimum parameters include F of about $\frac{1}{4}\gamma$, quite close to the above condition of significance. The particle's lagging the plane wave is thus about as important as the speeding up of the wave, and the practicable energy gain is correspondingly decreased.

These results indicate that at present and in the foreseeable future laser accelerators cannot compete with the higher-energy accelerators. But pulsed lasers are a simple, easy, and versatile means to impart large amounts of energy to charged particles, and might be useful in many specific applications. That peak laser powers can be expected to increase in the future makes this means of acceleration more interesting as time goes on.

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PHYSICAL REVIEW A

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Effects of a Volume-Dependent Potential on Equilibrium Properties of Liquid Sodium*

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Calculations of cohesive energy, pressure, and compressibility have been made for liquid Na at 393 °K. They are based on the Ashcroft form for the pseudopotential and the self-consistent dielectric function recently given by Singwi *et al.*, which have been found to give good agreement with the measured phonon dispersion relations in crystalline Na, and a liquid structure derived from a recent molecular-dynamics calculation by Rahman. The theory is formulated in terms of a volume-dependent pair potential and additional volume-dependent terms. A model which considers only volume-independent pairwise interactions gives incorrect results for the cohesive energy and pressure. It also gives a different result for the compressibility from the full theory, owing to the neglect of certain three- and four-particle interactions which are considered in the full theory; this result is, however, in reasonable agreement with experiment. The full theory gives good agreement for the cohesive energy, pressure, and compressibility if and only if an adjustment is made to the Hartree energy, similar to that made in a previous calculation for the crystal.

I. INTRODUCTION

The dynamics of metals in the solid and liquid state is often treated by assuming pairwise interactions between the ions. The validity of this approach is to some extent justified by the basic theory of simple metals.¹ If (a) the adiabatic approximation, and (b) perturbation theory to second order in the electron-ion interaction, are both valid, one can indeed define an effective potential between each pair of ions. The pairwise interactions, however, do not give a complete description of the system; not only are the effective pair potentials themselves volume dependent, but also the energy of the system contains additional terms depending on the volume alone. These features have an influence on the dynamic as well as the static properties; for example, they are primarily responsible for the failure of the Cauchy relations in metal crystals, as pointed out by Mott and Jones.² Furthermore, third- and fourth-order terms in the electron-ion interaction are probably not negligible even in the alkali metals; such terms represent three- and four-particle interactions in real space. In the long-wavelength limit some of these terms contribute to second order,^{3,4} and explicit calculations for the solid^{4,5} have shown that they are relatively

large.

On the other hand, the complexity of the problem of liquid dynamics is such that the assumption of pairwise interactions is almost indispensable. Models based on this assumption have been used in all molecular-dynamics⁶ and many-body theory⁷ calculations up to now. The principal aim of the present paper is to investigate the meaning of equilibrium properties such as cohesive energy, pressure, and compressibility for a model considering only volume-independent pairwise interactions (VIPI model). This is done by calculating both the pairwise interaction terms and the volume-dependent terms for liquid sodium. The calculations are based on a local pseudopotential and a dielectric function which give phonon dispersion relations in the crystal in very good agreement with those measured by neutron scattering, as described in an earlier paper,⁵ hereafter referred to as I. An effective pair potential (EPP) derived from these (Ref. 8, hereafter referred to as II) has been used in a molecular-dynamics machine calculation,⁹ and the pair distribution function (PDF) from that calculation has been used here. In this way one is able to make a completely consistent calculation with a realistic potential, avoiding the use of approximations like the hard-sphere model, which has been