Cluster Plasma and its Dispersion Relation

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February 13, 1998

Abstract

It is shown that unlike a gas plasma or an electron plasma in a metal, an ionized clustered material ("cluster plasma") permits propagation below the plasma cut-off of electromagnetic (EM) waves whose phase velocity is close to but below the speed of light. Its unique properties allow a variety of applications, including direct acceleration of particles with its EM fields and the phase matching of waves of high harmonic generation (HHG).

The properties of clustered materials have attracted interest for a long time (e.g. [1], [2]). Recently their interest has heightened, in part, due to the technical developments of nanotechnology and nanoparticles (e.g. [3],[4]) and, in part, due to irradiation of ultrashort laser pulses of recent times (e.g. [5]-[8]). For example, in the latter category of applications it has been shown that a cluster plasma can emit much more intense multiple harmonics X-rays than a comparable gas plasma upon irradiation of intense short laser pulses [9]. In these applications the clustered material is (highly or at the very least partially) ionized to form a cluster plasma. In spite of extensive theoretical studies (e.g. [1], [10]-[12]), to the best of our knowledge, we have not seen investigations of optical properties below the plasma cutoff in a cluster plasma. In this Letter we point out that electromagnetic (EM) waves can propagate in a cluster plasma below the plasma cutoff and describe their associated properties. In particular we apply these properties to suggest a novel particle acceleration method, without using an induced longitudinal field [13], and a phase matching method of waves of different harmonics generated by the mechanism of HHG (due to the mechanism ofthe optical periodic bremsstrahlung).

Consider, for the moment, a plasma of fully ionized clusters with the uniform electron density equal to all clusters. Let p be the packing ratio, i.e. the ratio of the volume occupied by all the clusters to the entire volume. Since our interest is focused on short pulse laser irradiation, ions in clusters are assumed to be immobile. A linearly polarized (in the y-direction) plane electromagnetic wave with frequency ω propagating in the x-direction is injected into this plasma. In this work we are interested in linear properties. We assume that the wavelength of this electromagnetic wave $\lambda = 2\pi/k$ is much greater than the (typical) cluster radius a. The Maxwell equation is

$$\frac{\partial^2}{\partial x^2} E_y - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} E_y = -\frac{4\pi}{c^2} \frac{\partial}{\partial t} \langle J_y \rangle, \tag{1}$$

where the current density $\langle J_y \rangle$ is average over the spatial wavelength of the electromagnetic

wave and

$$\langle J_y \rangle = -\langle n_e \rangle e \dot{y}, \tag{2}$$

where $\langle n_e \rangle$ is the average electron density and may be written as $\langle n_e \rangle = p\, n_e$, n_e is the local electron density inside of clusters ($n_e = Z\, n_i$, where n is the immobile ionic density). As we mentioned in the above, n_e is assumed to be uniform inside a cluster and has an equal value for all clusters (for now).

The dynamics of the electron displacement y is dictated by the electromagnetic field of course, but also now in a cluster, by the induced polarization on the cluster surface. The polarization in the direction (y) of the electric field of the EM wave in an ordinary plasma does not happen, though the polarization in the direction (x) of the propagation of the EM wave can happen in the intense regime (and has been exploited for acceleration [13]). However, for a cluster this situation fundamentally changes due to the surface charge induced polarization in the transverse (y)-direction. Because of this polarization, there arises a strong restoring force due to plasma electron space charge, which is different from but has similarity with the longitudinal plasma charge restoring force. The equation of motion of an electron in the y-direction, therefore, is

$$m\,\frac{d^2 y}{dt^2} + m f\, \omega_p^2 y = -e E_y(t), \tag{3}$$

where $\omega_p^2 = 4\pi e^2 n_e/m$, f is a geometrical factor of order unity (unity for slab clusters), and E_y is the laser electric field oscillating with frequency ω. Combining Eq. (3) and substituting the expression of y into Eq. (2), and then into Eq. (1), we obtain the dispersion relation of EM waves in a cluster plasma through the index of refraction $n(k\omega)$ as

$$n^2(k\omega) = \frac{c^2 k^2}{\omega^2} = 1 - \frac{p\omega_p^2}{\omega^2 - f\omega_p^2} - \left(\frac{q\,\omega_p^2}{\omega^2}\right), \tag{4}$$

where the last term in brackets may be added when there is a tenuous plasma background ($q \ll 1$). In the following we exclude the term in brackets for simplicity. (Inclusion of

these changes no qualitative properties of the dispersion relation, except one, which will be discussed later).

The dispersion relation of Eq. (4) has one fundamental difference from that for a gaseous (or metallic) plasma, i.e. the term in the denominator of the second term on the right-hand side, $-f\omega_p^2$ (see Fig. 1). This term arose from the transverse restoring force due to the space charge on the surface of clusters. Because of the presence of this term, Eq. (4) now yields two branches of dispersion relation and a new branch emerges that has a real frequency below the plasma cutoff, the two roots in a small wavenumber regime of Eq. (4) are:

$$\omega^2 \cong \begin{cases} (f+p)\omega_p^2 + \dfrac{\frac{1}{2}p}{f+p}\,c^2k^2, & \text{(5a)} \\[2ex] \dfrac{f}{f+p}\,c^2k^2, & \text{(5b)} \end{cases}$$

and those in a large wavenumber regime are:

$$\omega^2 \cong \begin{cases} c^2k^2 + p\omega_p^2, & \text{(6a)} \\[2ex] f\omega_p^2. & \text{(6b)} \end{cases}$$

Here both Eqs. (5a) and (6a) are the upper branch and correspond to the well-known EM wave dispersion relation in the presence of a plasma, while both Eqs. (5b) and (6b) emerge as a new lower branch. The most striking feature of this new branch is that the emergence of a propagating EM wave below the plasma cutoff (in the present case $\omega = \sqrt{f+p}\,\omega_p$). This branch has a phase velocity close to but less than the speed of light, $v_{ph} = c/\sqrt{(1+p/f)}$, and a resonance at $\omega = \sqrt{f}\,\omega_p$. Although in the present derivation no kinetic effect is included, it can be shown that this wave does not suffer from the Landau damping in the low k regime (even though $v_{ph} < c$), while it damps near the resonance due to the mechanism similar to cyclotron damping [14]. This is because the restoring force due to clusters does not allow ballistic motion to yield the Landau pole $(\omega - kv)^{-1}$. This situation is similar

to the Alfvén wave [15] in which the restoring force is the magnetic tension, where the phase velocity is typically $v_A \ll c$. In our dispersion, there is a narrow stop band between $\sqrt{f}\omega_p < \omega < \sqrt{f+p}\,\omega_p$. This is also similar to the theory of dielectrics [16].

Many applications can be derived from the properties of the new branch. Here we introduce a method of direct acceleration of particles. Let the EM wave (E_y, B_z) propagate in the x-direction. The equation of motion for a particle that propagates with the phase of the EM wave is

$$\frac{d\mathbf{p}}{dt} = -e\left[\widehat{y}\left(E_y - \frac{v_x B_z}{c}\right) - \widehat{x}\,\frac{v_y B_z}{c}\right], \tag{7}$$

where we set $E_y = v_x B_z/c$ to let the first term in Eq. (7) vanish, which can be written as

$$v_x = c\,\frac{E_y}{B_z} = \frac{\omega}{k_x} < c. \tag{8}$$

This last inequality in (8) arises from the property of the slow wave that emerged in the clustered plasma in Eq. (5b) and (6b). The satisfaction of Eq. (8) is not possible if the EM wave is in a gas plasma. Under Eqs. (7) and (8), it is, in principle, possible to continually accelerate electrons by keeping the condition Eq. (8) as long as this 1D plane wave is permitted. A similar continual acceleration with an imposed static magnetic field has been proposed [17]; the acceleration is sideways ("surfing"). In contrast the present method accelerates particles in the direction of laser propagation and the accelerating gradient is directly proportional to the laser magnetic field (in contrast to the static magnetic field in [17]), which allows a potential of realizing a large value of acceleration. An application of the current method to cylindrical geometry is of interest as well.

A variation of this particle acceleration is for pickup. As the density of electrons in cluster (thus ω_p) is decreased as a function of the propagation distance (x), the EM mode in the lower branch [L in Fig. 1(b)] marches up in the dispersion curve until it hits the resonance where the density gradually changes (an adiabatic change). See Fig. 1(b). On the other hand, when the electron density changes sufficiently quickly on x, the EM mode in

the laser branch [L in Fig. 1(b)] can tunnel through and mode-convert itself into one in the upper branch [H in Fig. 1(b)] (a non-adiabatic change). Since the frequency stop band of the cluster plasma is narrow, this mode-conversion is not difficult. The combination of the resonance, evanescence, and cutoff and propagation is the general property of the so-called Budden turning point [18]. Let us imagine a laser with frequency ω entering in a nonuniform cluster plasma. Here the nonuniformity may be determined or controlled by the local plasma density n_e (for example a different material or different ionization state) in a cluster or as the parameter f. Let us assume that the parameter $\sqrt{f}\, n_e$ is large enough so that the laser is propagating in the slow wave (the cluster mode) to begin with. Suppose that the parameter $\sqrt{f}\, n_e$ decreases as a function of the propagation distance. The laser EM wave will first encounter the resonance, then a cutoff. Because in our cluster plasma the gap is very narrow ($\propto p$), only over a very short interval the laser is evanescent, when it eventually enters the region of (fast wave, i.e. the ordinary) EM wave propagation. This constitutes a passage of the Budden turning point [18]. At the resonant point the wave phase velocity becomes much less than c, so that waves with enough amplitude can trap particles and then the phase velocity accelerates as it turns into the upper branch. This combination is thus ideal to pickup particles originally at rest and to accelerate them, which may offer an alternative to the "optical plasma cathode" [19]. It can be shown that with enough amplitude of EM waves, photon pressure helps to reduce the stop band barrier and to result in an enhanced transmission, which may be called self-indunced transmission (this nonlinear effect will be reported elsewhere).

Coming back to Eq. (4) with the last term (the coronal plasma), the dispersion relation at $\omega/\omega_p \to 0$ yields a portion of the dispersion curve with $n^2 < 0$. See a dotted curve in Fig. 1(b). This particular dispersion relation permits the refractive index (n) equality

$$n(\omega) = n(m\omega), \qquad m = \text{integer} \tag{9}$$

to be satisfied for a selected m-th order harmonic generation process. An example is illustrated in Fig. 1(b) with two such points connected. The condition, Eq. (9), is equivalent to exact phase matching for m-th order HHG, i.e. the wavenumber mismatch $\Delta\mathbf{k} = m\mathbf{k}(\omega) - \mathbf{k}(m\omega) = 0$ between the $\mathbf{k}$ vector $m\mathbf{k}(\omega)$ of the m-th order nonlinear polarization wave and the $\mathbf{k}$ vector $\mathbf{k}(m\omega)$ of the generated m-th harmonic EM wave. Condition (9) cannot be satisfied for a normally dispersive medium, such as a uniform plasma, in which $n(\omega)$ increases monotonically with ω [see the broken curve Fig. 1(b)]. The phase matching method should be distinguished from other phase matching methods. The methods used widely in low-order nonlinear optics—which are based on e.g. birefringence of nonlinear crystals, anomalous VUV dispersion of nonlinear gases, or periodic solid state quasi-phase-matched structures—are fundamentally incompatible with the short output wavelength (because of absorption) and the high input intensity (because of damage or ionization) involved in HHG, especially HHG in plasmas. Phase matching of HHG in plasmas thus requires completely new approaches. One approach proposed recently by Milchberg *et al.* [20] exploits the geometric dispersion of a plasma waveguide to compensate the material dispersion of a plasma. Like the method proposed here, this approach avoids limitations set by atomic ionization, but unlike the current method, relies on generating the harmonic output in a higher order transverse mode of the waveguide and is unfavorable for a direct $\omega \to m\omega$ harmonic generation process. Thus this approach has mainly been discussed in the context of high-order difference frequency processes—e.g. $\omega \to m\omega - m'\omega$. The current method, on the other hand, is compatible with lowest order Gaussian input and output modes and with direct $\omega \to m\omega$ harmonic generation.

Nevertheless, the feasibility of this HHG phase-matching method in the laboratory will depend on the sensitivity of the phase matching condition (9) to nonideal features of the nano-clustered plasma. These features include: 1. Nonuniformities in cluster size, packing fraction p, and coronal density $n_{ec} = q\, n_e$, which will affect the linear dispersion relation

(4) and the sharpness of the phase-matching condition (9). To estimate this effect, we have plotted (Fig. 2) the variation of the coherence length $L = \pi/|\Delta\mathbf{k}|$ as a function of the cluster packing ratio p and the coronal plasma density n_{ec} for 10th and 50th harmonic generation of a Ti:sapphire laser pulse in a clustered plasma described by Eq. (4). The sharp peak in these curves represents a perfectly phase matched condition. Since practical sample jets will have thicknesses $10^{-4}\text{m} < L_{\text{jet}} < 10^{-3}\text{m}$, the criterion $L \geq L_{\text{jet}}$ defines the tolerable p and n_{ec} ranges for these HHG processes to be effectively phase-matched. For these examples, experimentally feasible variations of several percent in p and n_{ec} are evidently tolerable. For applications in which lower conversion efficiencies, and therefore imperfect phase-matching, are acceptable, correspondingly greater nonuniformities will be tolerable. 2. Residual bound electron effects, such as d electrons in transition metals and surface resonances, which will alter the linear dispersion relation from the ideal fully ionized plasma form described by Eq. (4). The phase matching condition (9) is also sensitive to harmonic order m, thus providing a harmonic selectivity lacking in current HHG experiments. Such selectivity can be useful in many spectroscopic (e.g. surface science) applications of high harmonics. As an example, Fig. 2(c) presents a plot of the variation in the phase-matching efficiency factor $F = \sin^2(|\Delta\mathbf{k}|L_{\text{jet}}/2)/(|\Delta\mathbf{k}|L_{\text{jet}}/2)^2$, based on Eq. (4), as a function of harmonic order for plasma conditions for which 10th and 20th HG of Ti:sapphire laser pulse are perfectly phase-matched.

It is important to emphasize that the current theory is linear and the nonlinear physics should play an important role in the interaction between th cluster plasma and laser and will be a focus of future work.

We acknowledge discussions with Prof. J. Keto on cluster formation and anomalous dispersion. The work was supported in part by the U.S. Department of Energy and the National Science Foundation.

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FIGURE CAPTIONS

FIG. 1. The dispersion relations of the cluster plasma. (a) frequency ω vs. wavenumber k; (b) The dispersion relations [the two branches shown in (a) without the corona] along with the dispersion relation that includes the background diffuse (coronal) plasma [the term in brackets in Eq. (4)]. In this new dispersion two black points indicate the phase matching condition in Eq. (9).

FIG. 2. Variation in coherence length L as a function of (a) the cluster packing ratio p and (b) the coronal plasma density n_{ec} for the 10th and 50th harmonics of a Ti:sapphire laser ($\lambda = 800\,\mathrm{nm}$), based on Eq. (4). Unless varied, the cluster plasma parameters are: $f = 1$, $p = 0.01$, $n_{ec} = 2 \times 10^{10}\mathrm{cm}^{-3}$, and the cluster plasma density $n_{e\alpha} = 1 \times 10^{22}\mathrm{cm}^{-4}$; (c) variation in phase-matching efficiency $F = \sin^2(|\Delta\mathbf{k}|L_{\mathrm{jet}}/2)/(|\Delta\mathbf{k}|L_{\mathrm{jet}}/2)^2$ as a function of harmonic order for a Ti:sapphire laser. For the solid line, $t = 1$, $p = 10^{-2}$, $L_{\mathrm{jet}} = 0.2\,\mathrm{mm}$, $n_{ec} = 2 \times 10^{19}\mathrm{cm}^{-3}$, and $n_{e\alpha} = 1 \times 10^{22}\mathrm{cm}^{-3}$; for the dashed line, $i = 1$, $p = 10^{-4}$, $L = 2\,\mathrm{mm}$, $n_{ec} = 2 \times 10^{17}\mathrm{cm}^{-3}$, and $n_{e\alpha} = 1 \times 10^{23}\mathrm{cm}^{-3}$.