Simulation of discharging dust grains by laser excitation of neutral atoms

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We have simulated a method for changing the charge on dust grains in plasmas by exciting a small fraction of the neutral atoms into highly excited states. The atoms can be preferentially excited near a dust grain and quickly ionized by the hot electrons in the plasma. Because the neutral atom has low speed when it is excited, the resulting positive ion strikes the dust with nearly unit efficiency and the electron is repelled into the plasma. The rate for this process can be controlled by varying the state excited and/or the density of excited atoms. Thus, this mechanism gives a controllable method for varying the dust charge without substantially changing the other plasma properties.

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I. INTRODUCTION

The charge on a dust grain in a plasma depends^{1,2} on the size of the grain and the parameters of the plasma: electron temperature, ion temperature, mass of the ion, the ion and electron masses, and the local ion and electron plasma densities. However, the confinement^{3,4} and transport of a charged dust grain in the plasma depends critically on its charge. Also, the interesting collective phenomena (such as plasma crystals^{5,6} and Coulomb clusters⁷) present in dusty plasmas depend on the charge of the dust grain. Thus, there is a strong coupling between the dust grains and the surrounding plasma and what type of behavior is displayed. It would be of great interest to develop a technique that would allow the charge on the grain to be changed without changing the parameters of the plasma in which it is immersed, since even many of the in situ diagnostic techniques can perturb a dusty plasma.^{8,9}

Techniques for altering the dust grain charge without modifying the surrounding plasma have been considered for many years. From studies of astrophysical dusty plasmas, it is generally accepted that ionizing radiation (e.g., ultraviolet light) can cause the emission of electrons through the photoelectric effect.^{10,11} Recently, Land and Goedheer¹² performed one-dimensional Monte Carlo simulations of a dusty plasma that showed that for micron-sized grains, UV light could be effective at controlling the grain charge; however, detailed experiments have yet to be performed.

In this paper, we present an alternate method for controlling the charge on a dust grain by exciting atoms to a highly excited state when they are near the grain. A highly excited atom can be quickly ionized in a plasma from collisions with hot electrons. For example, an atom excited to a principle quantum number n=30 in a plasma with an electron density 10^8 cm⁻³ and electron temperature of 1 eV has a 1/e ionization lifetime of ~20 ns; if the atom has a speed of 400 m/s, it will travel less than ~8 μ m before being ionized. In typical laboratory dusty plasmas, micron-sized dust grains acquire a net negative charge of several hundred to several thousand electrons due to the higher mobility of the electrons compared to the ions. Thus, if the atom can be preferentially excited near the dust grain, it will be ionized close to the dust grain and the positive charge will be quickly pulled into the grain. Also, highly excited atoms can be stripped of the weakly bound electron by electric fields that depend on the size of the binding energy. For atoms in a state with principle quantum number *n*, the field to strip the electron is approximately $F_{\text{strip}} \sim 3.2 \times 10^8 \text{ V/cm/}n^4$. The field rapidly decreases with increasing *n*; this field is ~2 kV/cm for *n* = 20 and is ~400 V/cm for *n*=30. Typical electric fields in a plasma are a couple V/cm, but a 1 μ m radius dust grain with a charge of 2000 electrons has an electric field of approximately 2 kV/cm at a distance of 3.8 μ m from the dust center; the exact field strength depends on the Debye screening length. The field ionization of ground state atoms near a small structure (carbon nanotube) has been seen in Ref. 13.

The basic mechanism we propose for exciting the atom near the dust grain is the Stark shift of the excited state¹⁴ from the strong electric field near the dust. The deeply bound states of the atom hardly shift in energy but the highly excited states have polarizabilities that strongly increase with the principle quantum number, *n*: the polarizability scales such as n^7 . The shift of the excited energy level is $\Delta E =$ $-(1/2)\alpha F^2$ where α is the polarizability and *F* is the electric field strength; by detuning a laser from the vacuum energy, the atom will only be excited when it is near a region of space where the electric field has the value $F_a = \sqrt{-2\Delta E/\alpha}$.

Figure 1 gives a schematic of how the method proposed in this paper can work. An Ar atom with a kinetic energy of a few 100 K will essentially move with constant velocity until it reaches a position where the electric field is sufficient to satisfy the resonant condition. When the atom reaches such a field, it can be excited (Ar^*) by a laser and quickly thereafter is ionized by electron collisions or by the electric field from the dust that can be large if the atom gets close. Once the electron is stripped from the atom it moves radially outward into the plasma and the positive charge is trapped in the vicinity of the dust grain. This motion should be contrasted with that for a positive ion. The positive ion accelerates as it approaches the dust grain that means that it can easily miss a dust grain of radius 1 μ m if it reaches a radius of 10 μ m. However, this is not the case for an atom stripped at 10 μ m. For example, suppose the dust charge is 2000

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FIG. 1. A schematic sketch of the procedure described in this paper. An Ar atom moves in a straight line. The Stark shift of a highly excited state (Ar^{*}) is used to preferentially excite an atom at a distance R_a from the center of the dust grain. After traveling a short distance, the atom is ionized from a collision with a hot electron from the plasma. The high charge on the dust grain causes the electron to be repelled into the plasma and the positive ion to be attracted into the dust. The net effect is a decrease in the magnitude of the dust charge.

electrons, then the positive ion has a potential energy of approximately -3300 K and a kinetic energy of a few 100 K so it is bound to the dust grain and will hit it within a short time.

The excitation of the atom near the dust is the complicated aspect of the scheme we are proposing. This involves the physics of highly excited atoms in laser fields and electric fields. In particular, the important question is how many atoms are excited when they pass through fields that are changing with position.

We first give a simple theory to estimate the effect on the charge of the dust grain when electrons, positive ions, and highly excited atoms are present. This allows us to estimate the population of excited atoms that will be needed. We next describe how the atoms are excited when they reach the correct radius. We discuss some of the general trends and scaling for this system.

II. CHARGE BALANCE

In this section, we give a simplified treatment of the charge balance on a dust grain when electrons, positive ions, and highly excited atoms are present. This treatment includes the main effects in dust charging when the dust density is low and should be sufficient to indicate what density of excited atoms will be needed to see a substantial change in dust charge. The equations in this section are essentially the same as for the orbit motion limited approach and, thus, have the limitations of that method. However, the process described below is taking place at a scale length much greater than the dust radius but much less than both the ion and electron Debye lengths. Since the screening from the plasma surrounding the dust grain will be minimal, the ions created by stripping the electron from the highly excited atom will travel in a nearly ballistic trajectory to the grain. For the parameters we consider below, our estimates should accurately reproduce the trends in the charge as a function of the number of excited atoms (Debye length of ~120 μ m and excitation radius between ~20 and -28 μ m); when the distance, *r*, is much less than the Debye length, λ_D , the fractional change in the electric field is proportional to $(r/\lambda_D)^2$. To get the results of higher accuracy, a fully self-consistent calculation that includes screening effects should be performed; these effects will be more important for larger dust grains that have larger charge and, therefore, a larger distance from the dust where the excitation takes place.

For this derivation, we will assume that the dust grain is spherical with radius *R* and uniformly charged, the Debye length is much larger than the radius of the dust, and the mean free path of the charged particles is much larger than the dust radius. We will define the charge on the dust grain in terms of the electron charge to be $-Z_d e$ where $e=1.602 \times 10^{-19}$ C and Z_d is the excess number of electrons.

First, we will obtain an expression for the rate that electrons hit the dust grain. Only electrons with kinetic energy $KE \ge Z_d e^2/(4\pi\epsilon_0 R)$ can hit the dust grain. Because the dust grain is much more massive than the electrons and with much lower speed, we can approximate the grain as being fixed in space. In this case, electrons hit the grain when the impact parameter is less than

$$b^2 = R^2 \left(1 - \frac{PE_0}{KE} \right),\tag{1}$$

where $PE_0 = Z_d e^2 / (4\pi\epsilon_0 R)$. The collision cross section is $\sigma = \pi b^2$. Thus, the rate that electrons hit the grain is

$$\Gamma_e(v) = n_e \sigma v = n_e \sigma_0 \left(1 - \frac{PE_0}{KE} \right) v,$$
⁽²⁾

where $\sigma_0 = \pi R^2$ is the geometric cross section of the dust grain. If the electrons have a Maxwell–Boltzman distribution, then the average rate for hitting the grain is

$$\Gamma_e = 4\pi \left(\frac{m_e}{2\pi k_B T_e}\right)^{3/2} \int \Gamma_e(v) v^2 e^{-KE/k_B T_e} dv, \qquad (3)$$

where T_e is the electron temperature, m_e is the electron mass, k_B is the Boltzmann's constant, and the integration is only carried out over speeds where $\Gamma_e(v) \ge 0$. The integration can be analytically performed and the collision rate is

$$\Gamma_e = n_e \sigma_0 \sqrt{\frac{k_B T_e}{m_e}} \sqrt{\frac{8}{\pi}} e^{-P E_0 / k_B T_e}.$$
(4)

For a fixed PE_0 , the collision rate increases with increasing T_e . In practice, the charge on the dust depends on T_e and thus PE_0 can strongly depend on the electron temperature.

A similar treatment can be done for the positive ions. Unlike the electrons, the ions can hit the grain even when $v \rightarrow 0$. The ion collision rate is

$$\Gamma_i = n_i \sigma_0 \sqrt{\frac{k_B T_i}{m_i}} \sqrt{\frac{8}{\pi}} \left(1 + \frac{P E_0}{k_B T_i} \right), \tag{5}$$

where n_i is the ion density, T_i is the ion temperature, and m_i is the ion mass. For many situations, the $PE_0 \gg k_B T_i$ that will give the collision rate scaling with ion temperature such as $T_i^{-1/2}$. Thus, colder ions will lead to larger collision rates.

The collision rate for the atoms to be excited is simply the rate the atoms hit the radius, R_a , where the photon is absorbed. The electric field where the photon is absorbed is defined as F_a . Assuming that all of these atoms lead to a collision with the dust gives a collision rate

$$\Gamma_a = n_a \sigma_0 \sqrt{\frac{k_B T_a}{m_a}} \sqrt{\frac{8}{\pi} \frac{R_a^2}{R_0^2}},\tag{6}$$

where n_a is the density of *excited atoms*, T_a is the atom's temperature, and m_a is the atom's mass. Note that the collision rate scales with square of the excitation radius, R_a . However, this cannot be arbitrarily large because atoms that are ionized too far from the dust will not be captured and hit the dust. Also, we will see below that there is a trade-off in the excitation probability with R_a . The excitation radius depends on the charge of the dust grain and the excited state of the atom. Using the excitation electric field defined above, the excitation radius is

$$R_a \simeq \sqrt{\frac{e}{4\pi\epsilon_0 F_a}} \sqrt{Z_d},\tag{7}$$

where F_a is the electric field where the Stark shift puts the laser transition into resonance and $-Z_d e$ is the charge of the dust.

The number of excess electrons on a dust grain, Z_d , is found by having the electron collision rate equal to the sum of the ion and excited atom collision rate

$$\Gamma_e = \Gamma_i + \Gamma_a,\tag{8}$$

where the charge on the dust grain is only in the terms with $PE_0=Z_d e^2/(4\pi\epsilon_0 R)$ and in the excited atom collision rate because the stripping radius is proportional to $Z_d^{1/2}$.

In the discussion above, n_a is the density of *excited* atoms. This distinction is important because not all atoms that reach the excitation radius will be excited by the laser. In most dusty plasma experiments, the neutral density can be several orders of magnitude larger than the ion density. As will be seen below, large effects can occur when n_a is comparable to the ion density. Thus, only a very small fraction of the neutrals will need to be excited. The vast majority of laboratory dusty plasma experiments are performed in either dc or rf glow discharge plasmas with neutral pressures ranging from 0.1 to 30 Pa and ion densities $\sim 10^{14}$ to 10^{15} m⁻³.^{15,16} Therefore, in these experiments, the neutral density is $\sim 10^7$ times that of the ion density; in this case, less than 1 in a million atoms will need to undergo transitions to highly excited states.



FIG. 2. The number of excess electrons, Z_d , on a dust grain as a function of the ratio of highly excited atoms to ions. The plasma parameters and the dust radius are given in the text.

A. Example results

In this section, we will examine how the charge of the grain varies with the density of excited atoms for an example dusty plasma. In this example, we will take the dust to have a radius of 1.0 μ m, the electrons have a temperature of 1 eV, the ions and atoms to have a temperature of 300 K, and the atoms and ions having the mass of an Ar atom. We will assume the electron and ion density are the same. The dust charge will only depend on the ratio of the excited atom density to the ion density and on the electric field chosen for the laser transition. For this section, we will take $F_a = 35$ V/cm.

Figure 2 shows the charge on the dust grain as a function of the ratio of the excited atom density to the ion density. The charge is a monotonically decreasing function of the ratio: as the number of excited atoms increases the charge on the dust grain decreases. An important point is that the slope decreases with increasing n_a because the electrons can more easily hit the dust as the charge decreases. For the range of parameters shown, the charge on the dust decreases by a factor of ~2 due to the presence of the highly excited atoms.

There are a couple of scaling relations that arise from the simplified treatment of this section. The scalings should not be thought of as exact relations but as indicating the trends of how the charge changes with various parameters. For example, if both the radius, R, and the ratio n_a/n_i are decreased by the same factor, C, then the charge on the dust grain will also be decreased by the factor C. This means that if the radius is ten times smaller (0.1 μ m), then n_a/n_i needs to be 0.1 for a factor of ~ 2 decrease in the charge on the dust grain. Thus, the effect on the charge is larger when the dust grain has a smaller radius. Another important scaling relation is that the charge of the dust grain depends on the combination of excited atom density divided by the electric field, F_a , where the atom is excited. Thus, increasing F_a from 35 to 70 V/cm with a simultaneous increase in excited atom density by a factor of 2 will give the same effect.

III. ATOMIC TRANSITION

The transition to highly excited states of atoms has been accomplished many times over in different laboratories. This section examines the details of how this can be done.

Often, the highly excited states are reached by using more than one photon. For example, if the initial state is the Ar $3s^23p^54s$ J=2 metastable state, then a photon of ~4.19 eV is needed to directly reach highly excited states near n=25. However, these states can also be reached by first exciting to the $3s^23p^55p$ J=1 state using a 2.916 eV photon and then using a second photon with energy ~1.27 eV. These photons are relatively easy to produce. The 2.916 eV laser does not need to be very strong because this is a strong, dipole allowed transition. The 1.27 eV laser needs to be stronger because the dipole transition matrix element to highly excited states scales such as $1/n^{3/2}$. The only danger with this system is that the 2.916 eV laser depletes the $3s^23p^54s$ J=2 state.

The laser that is to cause the transition to the highly excited state is detuned so that the atom is only resonant when it is in the appropriate strength electric field. In this picture, the energy of the highly excited state is changing with time. Thus, the effective detuning of the laser is changing with time. To compute the size of the transition, we numerically solved the optical Bloch equations¹⁷ using a time dependent energy for the upper state; we used the rotating wave approximation for the interaction between the ground and Rydberg state.

Depending on the target state to be excited, the detuning cannot be arbitrarily chosen. There are two important considerations. The detuning should be substantially larger than the Doppler width of the line; the reason for this is that we are proposing to excite only near the dust and we do not want atoms in random places in the plasma to be excited because they are Doppler shifted into resonance. The Doppler energy width (full width at half maximum) is

$$\Delta E_{\text{Doppler}} = \sqrt{\frac{8k_B T \ln 2}{Mc^2}} E_0,$$
(9)

where E_0 is the transition energy. Taking T=300 K and E_0 = 1.27 eV gives a Doppler width of 2.5 μ eV. Thus, a detuning greater than ~25 μ eV should be sufficient. This corresponds to a detuning of ~6 GHz. However, there is a competing effect that must be taken into account. The detuning cannot be larger than the spacing of the energy levels of the highly excited atom that is ~27.21 eV/ n^3 . This gives the condition that $n < (27.21 \text{ eV}/\Delta E)^{1/3}$. Typically, this condition is easy to satisfy, but it *must be* satisfied otherwise the electric field is doing more than shifting the state. A different way of stating this energy shift condition is that the electric field must be small enough so that the eigenstates from different *n* do not mix. This condition is that $F_a < 1.7 \times 10^9 \text{ V/cm}/n^5$.

The transition probability to the highly excited state has scaling properties that can be used to establish the trends with F_a and with n. If the laser is strong enough, then all atoms will transition to the highly excited state when the



FIG. 3. The probability for an atom to be in an excited state as a function of time relative to the time when the resonant condition is satisfied. The solid line is from numerically solving the optical Bloch equation. The dotted line is the analytic expression equation (10).

field at the atom increases through F_a . If the laser is not this strong, then the transition probability can be computed using the Landau–Zener formalism¹⁸

$$P = 1 - \exp\left(-2\pi \frac{(F_{\rm las}D/2)^2}{\hbar |dE/dt|}\right),\tag{10}$$

where F_{las} is the laser electric field, *D* is the transition dipole moment, and dE/dt is the time dependent variation of the upper state energy when the resonant condition is reached; the combination $F_{\text{las}}D/2$ equals $\hbar\Omega$ where Ω is the Rabi frequency. Figure 3 shows the probability for an atom to be in the excited state as a function of time when $dE/dt/(2\pi\hbar)=(10^9 \text{ Hz})^2$ and $\Omega/(2\pi)=(1/2)$ MHz (the reason for choosing these parameters is given below). The time t=0 is when the upper level is shifted into resonance. As can be seen, the population in the excited state increases from 0 only during a small time when the resonant condition is satisfied.

One important scaling is that *D* is proportional to $1/n^{3/2}$ (Ref. 14) that means the transition probability decreases like $1/n^3$ for a fixed laser power and dE/dt. The square of the laser electric field is proportional to the laser intensity. The time variation of the upper energy depends on the geometry and the detuning. For small detunings compared to the energy spacing 27.21 eV/ n^3 , the energy variation of the upper state is from the polarizability. The detuning $\Delta E = -(1/2) \alpha F_a^2$ that is proportional to $1/R_a^4$ where R_a is the distance where the electric field from the dust equals F_a . From this dependence, the energy variation

$$\left|\frac{dE}{dt}\right| = 4 \left|\Delta E \frac{dR_a/dt}{R_a}\right|,\tag{11}$$

where dR_a/dt is the velocity of the atom dotted into the unit vector from the atom to the dust center. The dE/dt is proportional to the speed of the atom and proportional to $1/R_a^5$. Since the transition probability decreases as |dE/dt| increases, there will be more transitions if the transition distance can be increased; doubling the transition distance will increase the transition rate by a factor of ~30. The transition probability can also be increased if the speed of the atom is less. However, this effect is canceled by the fact that the flux of atoms is proportional to their speed; thus, there is not a strong motivation for controlling the speed of the atoms due to this consideration.

A final condition is that we are assuming that once the atom is ionized, the positive ion will be trapped to the dust. This sets an upper limit on the distance from the dust that the atom can be excited. When the atom is ionized, the total energy of the ion is the original kinetic energy of the atom plus the potential energy from the interaction with the dust. Roughly, the total energy is

$$E = KE - \frac{Z_d e^2}{4\pi\epsilon_0 r_i},\tag{12}$$

where r_i is the distance to the dust center when the ionization occurs. To be captured by the dust, this energy needs to be negative. Most of the ions will be captured if the potential energy is a few times k_BT_a . This gives the rough condition that

$$r_i < \frac{Z_d e^2}{4\pi\epsilon_0 5k_B T_a}.$$
(13)

For the example of Z_d =2000 and T_a =300 K, this gives a limit on $r_i \sim 20 \ \mu$ m.

IV. CONSISTENCY CHECK

In this section, we show that all of the conditions are satisfied for a specific case. In this case, we will assume that we are exciting a state with n=30 at a detuning such that the electric field where the excitation occurs is $F_a=35$ V/cm. Assuming the electron and ion density is 10^8 cm⁻³ with electron and ion temperatures of 1 eV and 300 K gives a Debye length of ~120 μ m.

For n=30, the maximum electric field that can be applied before the states of different *n* start mixing is 1.7 $\times 10^9$ V/cm/ $n^5 \sim 70$ V/cm; thus, the electric field is small enough.

At the maximum ratio in Fig. 2, the excitation radius, R_a , is less than 20 μ m. For that charge and radius, the potential energy is ~-800 K that should be compared to thermal energy of 300 K for the atoms. Only atoms with kinetic energy larger than 800 K will escape the dust. If the atoms are from a Maxwell–Boltzmann distribution, approximately 90% are trapped to the vicinity close to the dust grain.

The check whether the energy shift is outside of the Doppler width depends on the polarizability of the state that will depend on the atom and the particular state excited. A conservative estimate of the shift when the electric field is $\sim 1/2$ needed to mix states of adjacent *n* is $\sim 1/10$ the energy spacing of the *n*-levels. Taking this as the value gives ~ 25 GHz for the shift. This is well above the 6 GHz obtained from the Doppler estimate.

Finally, we address what fraction of atoms can be made to transition to the highly excited state. For small transition probabilities, we can rewrite Eq. (10) using the Rabi frequency of the transition

$$P = 2\pi\hbar\Omega^2 / |dE/dt|, \qquad (14)$$

where $\hbar\Omega = F_{\text{las}}D/2$. As an example, Ref. 19 obtained $\Omega/(2\pi) = (1/2)$ MHz for a laser power of 3.4 kW/cm² for a transition to n=43 state. The state in our example is n=30 that means a laser power of $\sim (40/30)^3 \sim 2.4$ less (i.e., 1.4 kW/cm²) would give the same Rabi frequency. From the discussion about calculating $dE/dt/(2\pi\hbar)$, we obtain $\sim 10^{18}$ Hz² if we use $\Delta E/(2\pi\hbar) = 10$ GHz, $R_a = 22 \ \mu$ m, and $dR_a/dt = 500$ m/s. Combining all of these numbers gives an excitation probability of $\sim 10^{-5}$; see Fig. 3 for the time dependence of the upper state population. If the laser intensity is increased by an order of magnitude from 1.4 kW/cm² to 14 kW/cm² then the transition probability increases by an order of magnitude. Although this transition probability is small it should be large enough to substantially change the charge on the dust.

V. CONCLUSION

We have simulated a possible mechanism for changing the charge on a dust grain in a dusty plasma. The mechanism involves exciting a small fraction of the neutral gas to a highly excited state that acts to discharge the dust grain. We have examined the main aspects of this scheme and found that it should be within the current state of technology.

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