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Charge-Dipole-Dipole Interactions in Ultracold Gases

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Abstract: Charge-dipole interactions are very common interactions among atoms and molecules, especially materials that can emit light or contain free charges. The second-order charge-dipole interactions, proportional to $\frac{1}{R^4}$, are stronger than the second-order dipole-dipole interactions or van der Waals interactions, proportional to $\frac{1}{R^6}$, at longer distances. In reality, there is more than one atom or charge; therefore, we focus on few-body charge-dipole interactions, such as charge-dipole-dipole interactions. Laser cooling and trapping allow us to study such interactions with much higher precision. In this article, charge-dipole interactions will be investigated in ultracold gases. To increase the interaction strength, we excite the ultracold atoms to highly excited states, Rydberg states. Here, we treat one Rydberg atom as a dipole, the excited electron and the ion core are the two poles of an electric dipole. Specifically, we study charge-atom interactions in ultracold Rydberg gases.

Keywords: van der Waals interactions; dipole-dipole interactions; charge-dipole interactions; ion-atom interactions; Rydberg atoms

1. Introduction

Charge-atom interactions or ion-atom interactions have been studied for a few decades. Charge transfer between ions and neutral atoms has been studied [1,2]. The $\frac{1}{R^4}$ dependence ion-atom interactions has been investigated [3]. Negative ions and atom interactions have been used to study electron attachment [4], and the calculation was done through the Configuration Interaction (CI) [4,5]. Power law potential has been used to calculate the ultracold ion-atom interactions [2,6,7], and power law potential has been applied to interactions between ions and Rydberg atoms [8]. Long-range charge-quadrupole interactions have been calculated [9], and charge-atom interactions are experimentally studied [10]. Imaging of ions by ion-Rydberg atom interaction induced absorption has been studied [11]. Ultracold atoms and ion-trap trapped ions have been theoretically and experimentally investigated [12,13]. Configuration-interaction effects have been examined for electron scattering by atoms and ions [14]. Hylleraas-configuration-interaction has been applied to Li atom Be^+ ions [15]. Negative ions can be produced through charge-atom and atom-atom interactions [16]. Charge-atom interaction can be used to study charge capture to create highly charged hollow atoms [17]. Spin-orbital interactions have been studied in cold ion-atom systems [18]. Positron-atom interactions have been examined in Ref. [19,20] and references therein. Magnetically tunable interactions have been studied [21]. Rydberg atom-ion molecules are proposed [22].

This topic is also relevant to the applications of van der Waals interactions among highly excited states [23,24]. Van der Waals interactions, the second-order dipole-dipole interactions, are widely studied in ultracold gases, especially in ultracold Rydberg gases., for which the energy of Rydberg atoms is close to the ionization limit. In other words, Rydberg atoms are weakly bounded compared to lower energy atoms. Therefore, the electrons are easily ionized, and both the ion and the electron become free charges. Due to the fact that the electron is a lot lighter than ions, the electron will quickly leave the atomic sample due to the much higher speed of the electron than the positive ion according to momentum conservation if there are not enough ions accumulated. However, if there are enough positive ions accumulated in the atomic sample, the free electron will not be able to escape from the Coulomb force created by the positive ions. Therefore, plasma will be created at that point [25], and this is not a regime that we are interested in. In this article, we study the effects of free charges on neutral atoms. In other words, a few ions are among the highly excited Rydberg atoms, and there is no plasma formation in the sample. Most quantum systems involving Rydberg atoms are carefully designed,

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so this is not an issue. For new applications involving Rydberg atoms, this might be a concern at the early stage of the design process. In this article, we study the effects of free charges on Rydberg atoms. Specifically, we will be focusing on the charge-dipole-dipole interactions. Here, we treat one Rydberg atom as a dipole; an outer electron and an ion core are the two poles of an electric dipole. In addition, we add more atoms and study the effects between one charge and few-body interactions.

2. Theory

We apply the Born-Oppenheimer approximation; the electronic motion and the nuclear motion can be separated, and we focus on the electronic motion. The Hamiltonian of a system with one ion and multiple atoms is the following:

$$H = \sum_{i}^{N}(-\frac{\hbar^2}{2m_i}\nabla^2 - \frac{e^2}{4\pi\epsilon_0 r_i}) - \frac{\hbar^2}{2m_{ion}}\nabla^2 + V, \tag{1}$$

where the terms in the parentheses are the kinetic energy and potential energy of the i^{th} atom, and i indicates the i^{th} atom of the total N atoms. $-\frac{\hbar^2}{2m_{ion}}\nabla^2$ is the kinetic energy of the ion. V is the sum of all inter-atomic potential energies as well as the potential energies between the atoms and the ion. For example, if we consider two-body charge-dipole interactions, N=1, and

$$V = V_{cd} + V_{dc}. \tag{2}$$

V_{cd} and V_{dc} are charge-dipole interactions and dipole-charge interactions as explained below. In this case, we consider the exchange symmetry. In other words, we consider both configurations shown in Figure 1a,b.

We study the three-dimensional charge-multipole interactions. The three-dimensional charge-dipole interaction, V_{dc}, between atom 1 and charge 2 is

$$V_{dc} = \frac{ep_1}{4\pi\epsilon_0 R^2}[\frac{1}{\sqrt{2}}sin\theta e^{i\phi}C^1_{1,-1} - \frac{1}{\sqrt{2}}sin\theta e^{-i\phi}C^1_{1,1} + cos\theta C^1_{1,0}], \tag{3}$$

where $C_{k,q}$ is a spherical tensor [26], and the superscripts 1 and 2 in $C_{k,q}$ mean that the spherical tensor operator is acting on the first atom and the second atom respectively. θ is the angle between R and the z axis, and ϕ is the angle between x axis and the projection of R on $x-y$ plane as shown in Figure 1a. p_1 is the dipole moment of atom 1 in Figure 1a. In this article, we consider exchange symmetry. If we switch the position of the charge and the atom, we get the dipole-charge interactions, which have the following form

$$V_{cd} = -\frac{ep_2}{4\pi\epsilon_0 R^2}[\frac{1}{\sqrt{2}}sin\theta e^{i\phi}C^2_{1,-1} - \frac{1}{\sqrt{2}}sin\theta e^{-i\phi}C^2_{1,1} + cos\theta C^2_{1,0}], \tag{4}$$

where p_2 is the dipole moment of atom 2 shown in Figure 1b.

If there are multiple atoms, the atom-atom interactions can be included in the V term in Equation (1). For example, if $N = 2$, we consider the configuration shown in Figure 1d. In addition, we consider dipole-dipole interactions and lower-order interactions; we ignore higher-order charge-multipole and multipole-multipole interactions. The potential energy is

$$V_3 = V_{cd} + V_{dc} + V_{dd}. \tag{5}$$

where V_{cd} is the interaction potential energy between the top charge, charge 2, and the bottom-left atom, atom 1. V_{dc} is the interaction potential energy between the bottom-right atom, atom 3, and the top charge, charge 2. V_{dd} [27] is the interaction potential energy between the two atoms at the bottom of Figure 1d.

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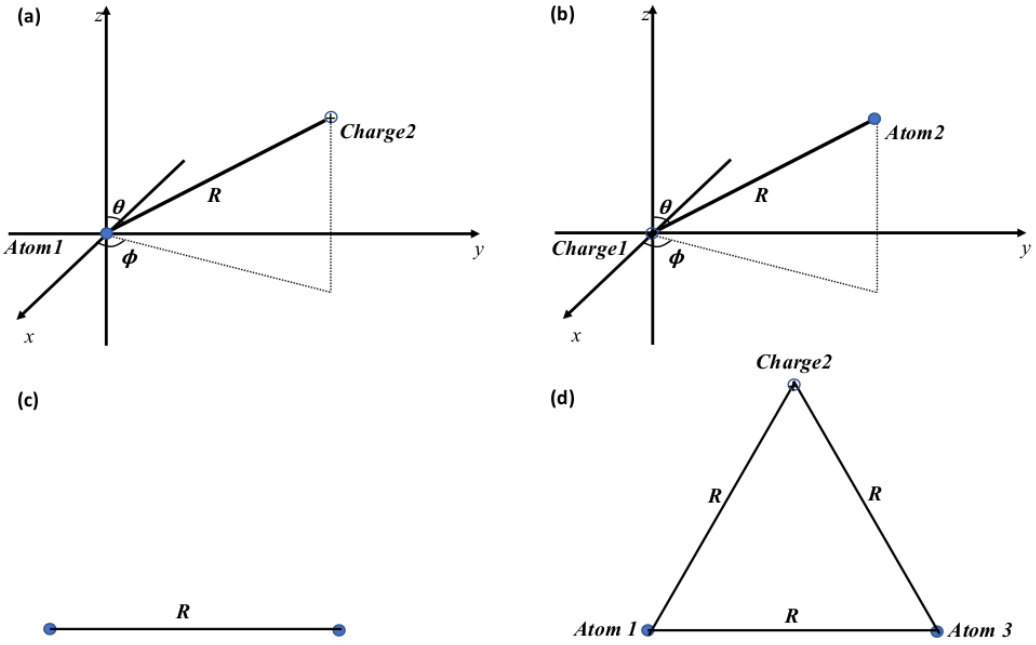

Figure 1. (a) Atom 1 is located at the origin, and the distance between atom 1 and charge 2 is R. (b) Charge 1 is located at the origin, and the distance between atom 2 and charge 1 is R. (c) Two atoms are separated by a distance R. (d) Two atoms, atom 1 and atom 3, and one charge, charge 2, are located at the three vertices of an equilateral triangle with a side length R.

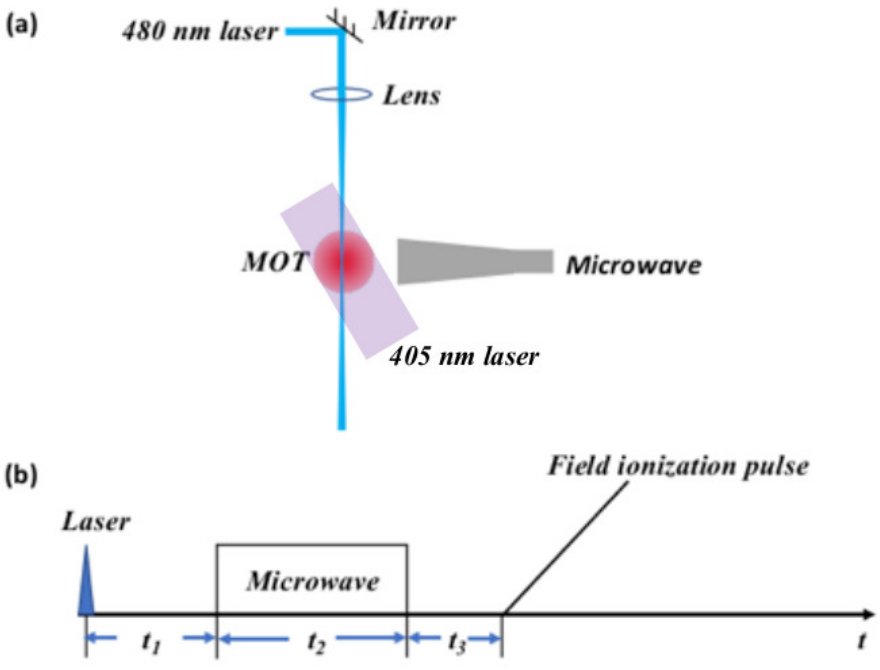

Figure 2. (a) The experimental setup. The MOT is held in a vacuum chamber with pressure less than 10^{-8} torr. (b) The timing of this experiment. In this experiment, $t_1 = 0.85$ μs, $t_2 = 1$ μs, and $t_3 = 0.65$ μs.

3. Experiment

In this experiment, we use a typical ^{85}Rb Magneto-Optical Trap (MOT) [28–34]. A 480 nm Nd:YAG laser-pumped pulsed dye laser is reflected from a mirror and then focused on the MOT through a

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lens to excite $5p_{3/2}$ atoms to a highly excited state, a Rydberg state. The free charges are created by a free-running CW (Continuous Wavelength) 405 nm diode laser with 100 mW power expanded to cover the whole MOT. The amount of ions added to the system is controlled by the amount of attenuation added to the diode laser. The kinetic energy of the free charges is about 0.5 eV. After waiting for about 850 ns after the Rydberg excitation, a 1 μs long microwave pulse is added, followed by a field ionization pulse [35] applied 650 ns later. The ionized free ions are collected by a pair of MCPs (Micro Channel Plates), which are connected to an oscilloscope. The oscilloscope signal is then imported by a computer and analyzed later.

4. Result and Discussion

Figure 3 shows two microwave scans taken for the 36*s* to 37*s* atomic transition. The black curve was taken without free ions, and the red curve was taken with free charges. It is known that due to the van der Waals interactions between atoms, the atoms are no longer isolated. The black curve slightly broadens in the lower frequency end caused by the van der Waals interactions. By adding free charges to the sample, the broadening is magnified, which indicates that by adding free charges, the van der Waals interactions between atoms are enhanced. Figure 4 shows the calculations of the interactions between 36s and 37s atoms with (the red curves) and without free charges (the black curves) shown in configuration Figure 1(d) and 1(c). The top group of states are the $36p_{3/2}36p_{3/2}$ states, and the bottom group of states are the 36s37s states. Both groups of states couple with van der Waals interactions. The Hamiltonian used for this calculation is shown in Equations (1) and (5). One GHz in Figure 3 is equivalent to 2 GHz in Figure 4 since the transition is a two-photon transition. In the experiment, we studied the bottom group of states shown in Figure 4. It is shown that by adding a charge to the system, the energy levels broaden more to the lower frequency side. In other words, the 36s to 37s transition spectrum line slightly broadens to the lower frequency side due to van der Waals interactions. By introducing free charges, the broadening caused by van der Waals interactions is enhanced. The experiment and the theory agree very well.

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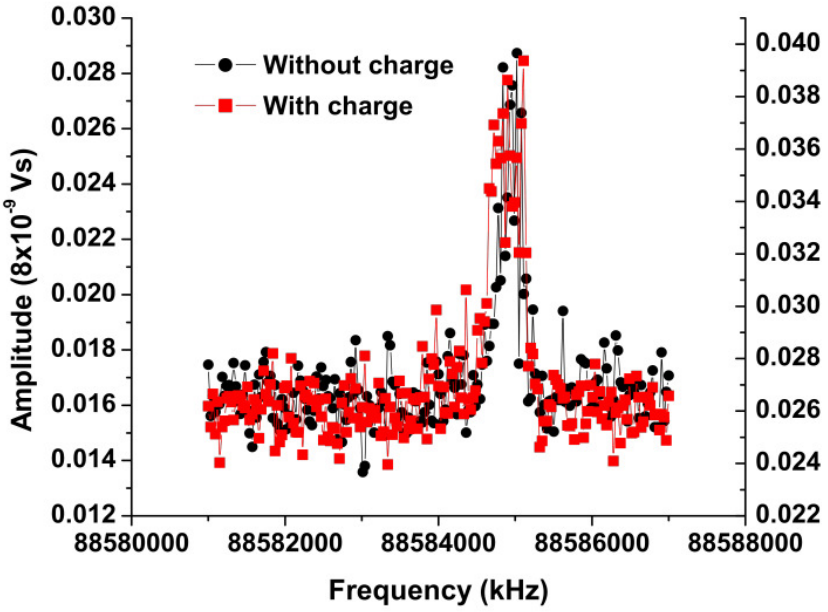

Figure 3. Two microwave scans were taken for the 36s37s state, which has attractive dipole-dipole interactions. The back curve (-■-) is taken without charges and the red curve (-●-) is taken with free charges. The horizontal axis is one microwave photon frequency of this two-photon transition. The vertical axis is proportional to the size of the Rydberg signal. It is calculated by integrating the corresponding peak of the field ionized signal on the oscilloscope. For example, 0.02 means 1.6×10^{-10} Vs (or Volts times Seconds).

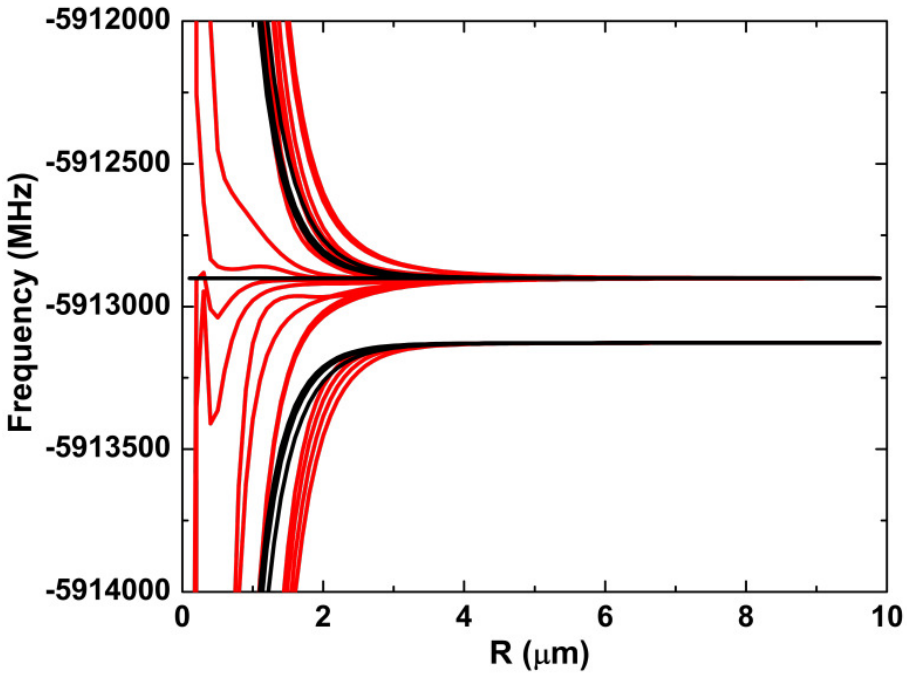

Figure 4. Calculated energy levels. The back curves (——) are calculated without charges, and the red curves (——) are calculated with free charges. The horizontal axis is the internuclear spacing between the two 36s and 37s atoms or the side length of the equilateral triangle shown in Figure 1(d), and the vertical axis represents the absolute energy of the energy levels divided by Planck's constant.

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5. Conclusions

In conclusion, charge-atom interactions are studied both theoretically and experimentally. It has been shown that by adding charges to the highly excited Rydberg atomic sample, the broadening caused by van der Waals interactions is enhanced. This has many applications. For example, charge-atom interactions can be used to create molecules more efficiently. In addition, Coulomb blockade can be investigated in this system. Moreover, since charge-atom interaction is the second strongest electromagnetic interaction at long distances, it can be used to study long-range entanglement, as well as quantum sensing.

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