Two-dimensional three-body quadrupole–quadrupole interactions

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Abstract

The interactions between dipoles are called dipole–dipole interactions. One type of dipole–dipole interaction is van der Waals interaction. Similar to van der Waals interactions, the interactions between quadrupoles are called quadrupole–quadrupole interactions. Atoms can be treated not only as dipoles but also as quadrupoles. In this article, we study the quadrupole interactions between highly excited atoms or Rydberg atoms. In addition, unlike many other calculations, in which the primary focus was on the one-dimensional two-body quadrupole–quadrupole interactions, the primary aim of this article is to study the two-dimensional few-body quadrupole–quadrupole interactions. Specifically, the two-dimensional three-body interactions are investigated. This research has many applications, such as quadrupole-blockade for quantum computing, creating molecules based on quadrupole interactions, or quadrupole–quadrupole molecules.

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1. Introduction

Quadrupole–quadrupole interactions have been studied in different areas of research, such as nuclear physics, ionic or plasma physics, and molecular physics. For example, quadrupole interactions between molecular rotation and a spinning nucleus have been investigated [1]. Quadrupole–quadrupole interactions between nucleons have been reported, which may give rise to a variety of collective effects [2]. The quadrupole–quadrupole interactions between nuclei and impurity paramagnetic ions in metals and alloys were indirectly analyzed through conduction electrons [3]. It has been shown that the quadrupole–quadrupole interactions play a crucial role in driving nuclear-excited states [4]. Moreover, density broadening has been studied in different disciplines. The quadrupolar interactions between two ions have been examined [5]. Quadrupole–quadrupole interactions have been used to study plasma-induced transparency [6]. Furthermore, quadrupole–quadrupole interactions contribute to the $C_5$ coefficient, which has been calculated for Cs [7]. Quadrupole–quadrupole interactions have been studied in molecules [8, 9]. The angular dependence of quadrupole–quadrupole interactions is investigated in ultracold gases [10]. However, most of the studies focus on two-body one-dimensional interactions. In this article, we focus on the three-dimensional few-body quadrupole–quadrupole interactions, and lower-order interactions, such as dipole–dipole interactions and dipole-quadrupole interactions among neutral atoms, are ignored. The on-resonance dipole–dipole interactions, which are proportional to $1/R^3$ [11], are negligible if all the atoms are in one state. The van der Waals interactions, which are proportional to $1/R^6$, are more likely to cause asymmetric broadening [12]. Dipole-quadrupole interactions involved in this case are proportional to $1/R^8$. On-resonance quadrupole–quadrupole interactions, which are proportional to $1/R^5$, are experimentally tested [13] in laser-cooled [14–20] highly excited states [21, 22]. Such interactions have a variety of applications,
such as quadrupole–quadrupole blockade for quantum gates. The experimental testing of the quadrupole blockade and related studies are very similar to the dipole-blockade [23–32]. These interactions can also be used for quantum information storage at higher densities and quadrupole–quadrupole coupled molecules. This work lays the foundation for many applications, such as the future generation of quantum computers and accurately calculating energy levels for molecules with different geometries.

This paper is arranged in the following way: the quadrupole–quadrupole interactions are presented in the next section, which is followed by the discussions about applications to different cases.

2. Theory

Quadrupole–quadrupole interactions can be calculated by expanding the Coulomb interactions between four charges [33]. In this article, we assume the four charges are from two atoms separated by a distance $R$ as shown in figure 1. We further assume that the internuclear spacing, $R$, is much greater than the size of the atoms, $r_1$ and $r_2$. The Born–Oppenheimer approximation is applied. Specifically, the positive ions have zero kinetic energy. The Hamiltonian for this system is

$$H = -\frac{\hbar}{2\mu_1} \nabla_1^2 - \frac{\hbar}{2\mu_2} \nabla_2^2 + V_1 + V_2 + V_{12},$$

(1)

where $\mu_1$ and $\mu_2$ are the effective mass of atom 1 and 2, and the bottom atom and the top atom. The first two terms are the kinetic energy of the two atoms. The third and fourth terms are the internal Coulomb potential energy within atom 1 and atom 2 respectively, $V_1 = -\frac{e^2}{4\pi\epsilon_0 r_1^2}$ and $V_2 = -\frac{e^2}{4\pi\epsilon_0 r_2^2}$. $V_{12}$ is the quadrupole–quadrupole interactions between four charges shown in figure 1, which can be written as:

$$V_{12} = \frac{Q_1 Q_2}{4\pi\epsilon_0 R^5} \left\{ \frac{35}{8} \sin^4 \theta \left( e^{i\phi} c_{1,2}^{(1)} c_{2,1}^{(2)} + e^{-i\phi} c_{1,2}^{(1)} c_{2,1}^{(2)} \right) + \frac{35}{8} \sin^2 \theta \sin(2\theta) \right.$$

$$\times \left[ e^{3i\phi} c_{2,1}^{(1)} c_{1,2}^{(2)} + c_{1,2}^{(1)} c_{2,1}^{(2)} \right]$$

$$- e^{-3i\phi} c_{2,1}^{(1)} c_{2,1}^{(2)} c_{2,1}^{(2)} + \left( 7 \cos^2 \theta - 1 \right)$$

$$\times \left[ \frac{5}{8} \sqrt{6} \sin^2 \theta \left( e^{-i\phi} c_{1,2}^{(1)} c_{2,0}^{(2)} + c_{1,2}^{(1)} c_{2,0}^{(2)} \right)$$

$$+ e^{2i\phi} c_{1,2}^{(1)} c_{2,0}^{(2)} + c_{1,2}^{(1)} c_{2,0}^{(2)} \right]$$

$$\left. + \frac{5}{8} \sin^2 \theta \left( e^{-2i\phi} c_{2,2}^{(1)} c_{2,2}^{(2)} + e^{2i\phi} c_{1,2}^{(1)} c_{2,2}^{(2)} \right) \right\}$$

$$+ \frac{7}{4} \sin^2 \theta - 1 \right) \frac{5}{8} \sin(2\theta) \left( e^{-i\phi} \left( c_{1,2}^{(1)} c_{2,1}^{(2)} + c_{1,2}^{(1)} c_{2,1}^{(2)} \right)$$

$$+ e^{-i\phi} \left( c_{1,2}^{(1)} c_{2,1}^{(2)} c_{2,1}^{(2)} + \right. \left. c_{1,2}^{(1)} c_{2,1}^{(2)} c_{2,1}^{(2)} \right)$$

$$+ (3 - 7 \cos^2 \theta) \frac{5}{8} \sqrt{6} \sin(2\theta) \left( e^{-i\phi} \left( c_{1,2}^{(1)} c_{2,1}^{(2)} \right.$$} + C_{1,2}^{(1)} c_{2,0}^{(2)} + C_{1,2}^{(1)} c_{2,0}^{(2)} \right)$$

$$+ C_{1,2}^{(1)} c_{2,0}^{(2)} + C_{1,2}^{(1)} c_{2,0}^{(2)} \right\},$$

(2)

where $Q_1 = e r_1^2$ and $Q_2 = e r_2^2$ are the quadrupole moments of atom 1 and atom 2. $R$ is the distance between the two atoms as shown in figure 1. $C_{2,0}$ is the second-order spherical tensor in Edmonds [34]. Here $q = -2, -1, 0, 1, 2$. It is shown that if $\theta = 0$ and $\phi = 0$, equation (2) reduces to the one-dimensional quadrupole–quadrupole interactions [10]. In addition, it can be proved that by reversing the $R$ direction shown in figure 1, or changing $\theta$ to $180^\circ - \theta$ and $\phi$ to $\phi + 180^\circ$, the $V_{12}$ remains the same. Unlike the one-dimensional quadrupole–quadrupole interactions, the $M$ or the projection of the total angular momentum of all three atoms in the $z$-axis, is no longer conserved in the three-dimensional quadrupole–quadrupole interactions. Therefore, the number of matrix elements is significantly increased for the three-dimensional quadrupole–quadrupole interactions compared to the one-dimensional quadrupole–quadrupole interactions.

For three-body quadrupole–quadrupole interactions, the Hamiltonian can be modified from equation (1) by adding an additional atom:

$$H = -\frac{\hbar}{2\mu_1} \nabla_1^2 - \frac{\hbar}{2\mu_2} \nabla_2^2 - \frac{\hbar}{2\mu_3} \nabla_3^2 + V_1 + V_2 + V_3 + V_{123},$$

(3)

where $-\frac{\hbar}{2\mu_1} \nabla_1$, $-\frac{\hbar}{2\mu_2} \nabla_2$, $-\frac{\hbar}{2\mu_3} \nabla_3$ are the kinetic and internal potential energy of atom 3. $V_{123}$ is the sum of all pair-wise potential energies between the three atoms:

$$V_{123} = V_{12} + V_{23} + V_{31},$$

(4)

Very similar to the quadrupole–quadrupole interactions between atom 1 and atom 2 shown in equation (2), $V_{23}$ is the quadrupole–quadrupole interaction potential energy between atom 2 and atom 3, and $V_{31}$ is the quadrupole–quadrupole interaction between atom 3 and atom 1.

3. Results and discussion

In this article, we consider two-dimensional three-atom interactions. Specifically, the ddd Rydberg states [21, 22] are considered. The initial state is $34d_{5/2,3/2,1/2}$ and $34d_{3/2,3/2,1/2}$. In other words, three $34d_{5/2,3/2,1/2}$ atoms are located at the three vertices of an equilateral triangle as shown in figure 2. The total number of elements considered is $(2 + 10)^3 = 123^3 = 1728$, since the number of m states in the s state is 2, and the total number m states in the d states is 10 (in the $d_{3/2}$ states and 6 in the $d_{5/2}$ states), where m is the projection of the total angular
Figure 1. Two ion cores of the two atoms, atom 1 (the bottom atom) and atom 2 (the top atom), are located at the origins of the two Cartesian coordinates, $O$ and $O'$. The distance between the bottom ion core and the electron of the bottom atom is $r_1$, and the distance between the top ion core and the electron of the top atom is $r_2$. The angle between $r_1$ and the $z$ axis is $\theta_1$, and the angle between the projection of $r_1$ in the $x$–$y$ plane and the $x$ axis is $\phi_1$. The distance between the two ion cores is $R$, the distance between the two electrons is $R'$, the distance between the ion core of the bottom atom and the electron of the top atom is $R''$, and the distance between the electron of the bottom atom and the ion core of the top atom is $R'''$.

Figure 2. Three atoms are located at the three vertices of an equilateral triangle, and each atom is composed of one ion core and one electron. The distance between two neighboring atoms is $R$.

Figure 3. The energy, or frequency, as a function of the internuclear spacing. The energy levels calculated for the $34d_3^12^1$ and the initial state is $34d_5^12^1$. On the right side of the figure from the top to the bottom, the levels are $34d_5^12^1$, $34d_5^12^1$, $34d_5^12^1$, and $34d_5^12^1$. In addition, the second level has three-fold degeneracy at $R \to \infty$, and the third level has three-fold degeneracy at $R \to \infty$. Similarly, the third level has three-fold degeneracy at $R \to \infty$, and the bottom level shows purely attractive quadrupole–quadrupole interactions, and some levels show both attractive and repulsive quadrupole–quadrupole interactions. There are still some internuclear spacings, and the level structure is sophisticated. However, there are some general features. For example, the bottom level shows purely attractive quadrupole–quadrupole interactions, and some levels show both attractive and repulsive quadrupole–quadrupole interactions. There are still some
open areas, which will be transparent for certain frequencies. Such a structure is very useful for creating transparent materials at a certain frequency range and block other frequencies. It is also shown that potential wells can be created through quadrupole–quadrupole interactions in a two-dimensional case. Moreover, potential peaks are observed. Those peaks can be as high as 100 THz or higher, which can possibly be used for quantum information storage.

Figure 4 shows the principal quantum number \( n \) dependence of the maximum frequency shift. Specifically, the maximum repulsive frequency shift at the internuclear spacing \( R = 0.5 \, \mu \text{m} \) vs \( n^8 \) is plotted. It is shown that the linear fit is very close. The reason is that the radius of the atoms is proportional to \( n^6 \), and each quadrupole moment has \( r^2 \) in it. Therefore, it is expected that the total shift is proportional to \( n^8 \).

4. Conclusion

In summary, the three-dimensional quadrupole–quadrupole interaction potential energy is expressed. In addition, this expression is applied to the three atoms located at three vertices of an equilateral triangle, and all three atoms are in the d states. Moreover, the principal quantum number \( n \) dependence is investigated. It is shown that the maximum repulsive frequency shift depends on \( n^8 \). This work lays the foundation for many applications, such as the future generation of quantum computers and accurately calculating energy levels for molecules with different geometries.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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