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To cite this article: B V Zelener *et al* 2016 *J. Phys.: Conf. Ser.* **774** 012163

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Thermodynamic of mixtures of ultracold Rydberg gases with different levels of excitation

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Abstract. The physical model of the particle system describing the thermodynamics of mixtures of ultracold Rydberg gases with different levels of excitation are proposed. In our model system of particles is a mixture of atoms in S -state and atoms in the P -state. To calculate the thermodynamic of the mixture Rydberg gases the method of Monte-Carlo was used. Computational results of the spatial correlations of dipoles depending on the coupling parameter are presented. At some value of the nonideality parameter the dipole chains are formed.

1. Introduction

The equation of state of highly excited gas in gas lasers (room temperature and atmospheric pressure), was first discussed in [1, 2]. These works have demonstrated the possibility of decay of the excited gas into two phases with different relative contents of excited atoms. This is due to the fact that the dipole-dipole interaction ($V_d \sim 1/r^3$) of two identical atoms with different energy levels averaged over all possible orientations does not disappear. This is possible only, if we assume that all orientations of the dipole moment have equal probability. It was shown [1, 2] that a more correct averaging, which takes into account the mutual correlation of the orientations leads to the fact that the main contribution to the sum of the states of the gas gives terms proportional r^{-3} . All the estimates in [1, 2] were made by the virial expansion under the condition of small number of excited atoms.

Our interest in studying the equation of state of highly excited gas is caused by the possibility of a phase transition in an ultracold gas of Rydberg atoms [3–7], the density of which is small, but the temperature is a few millikelvins. The possible numerical calculation of the thermodynamic functions for this physical model is the Monte-Carlo method.

So, the paper considers a physical model of a system of particles that describe the thermal dynamics of the mixture of ultracold Rydberg gases with different levels of excitation. In our model system of particles is a mixture of atoms N_S in S -state and atoms N_P in the P -state.

2. Quantum calculation of average dipole moment

We must take into account the fact that in the experiments [6, 7] not only the energy of the valence electron was fixed, but also its orbital angular momentum l . As a result of the transition $2P_{1/2(3/2)} \rightarrow nS_{1/2}$ to a highly excited state ${}^7\text{Li}$ formed a state with $l = 0$. In this case $N_P = 0$, so the only interaction is one between two nS atoms.



In order to calculate the probability of a certain value of the dipole moment projection let us present the state $nS_{1/2}$ as the combination of the parabolic eigenfunctions. The coefficients of the expansion of spherical functions $|n, l, m\rangle$ on a parabolic $|n_1, n_2, m\rangle$ ($n_1, n_2 \geq 0, n_1 + n_2 = n - |m| - 1$) are coefficients of the Clebsch-Gordan. This is due to the fact that the functions $|n_1, n_2, m\rangle$ are eigenfunctions of the two moments $\hat{\mathbf{j}}_1, \hat{\mathbf{j}}_2$ ($|n_1, n_2, m\rangle = |j_1, \mu_1\rangle |j_2, \mu_2\rangle$) and $j_{1z} = \frac{1}{2}(m + n_1 - n_2) = \mu_1, j_{2z} = \frac{1}{2}(m - n_1 + n_2) = \mu_2, j_1 = j_2 = \frac{n-1}{2}$ and $\mathbf{l} = \mathbf{j}_1 + \mathbf{j}_2$.

The function, which a decomposition we are interesting in, is the function $|n, 0, 0\rangle$ (state $nS_{1/2}$), have non-zero dipole moment directed along the axis z or in the opposite direction. The probability of a certain value of the dipole moment is determined by the square of the corresponding coefficient Clebsch-Gordan.

X_μ is a native vector of the matrix $\hat{\mathbf{I}}^2$ with eigenvalue 0. Non-zero components of this matrix $\mu - \mu' = \pm 1, 0$. The system of n homogeneous equations for X_μ has the form ($A_{-\frac{n-1}{2}} = C_{\frac{n-1}{2}} = 0$):

$$A_\mu X_{\mu-1} + B_\mu X_\mu + C_\mu X_{\mu+1} = 0. \quad (1)$$

The coefficients of the equations satisfy the equality $A_\mu + B_\mu + C_\mu = 0$. The solution of this system is the sequence of numbers equal in absolute value. Therefore, in the expansion of functions $|n, 0, 0\rangle$ by parabolic functions all of these functions are presented with an equal weight.

This in turn means that in the decomposition of functions $|n, 0, 0\rangle$ by parabolic functions $|n_1, n_2, 0\rangle$ they are all presented with an equal weight. The average value of $d = ez$ in this state (see for example [8]) is:

$$\langle d \rangle = \frac{6\eta^2}{me}(n_1^2 - n_2^2). \quad (2)$$

Believing $n_1 = k, n_2 = n - 1 - k, k \geq (n - 1)/2$, we have:

$$\langle d \rangle = \frac{6\eta^2}{me}(2k(n - 1) - (n - 1)^2), \quad d_{\max} = \frac{6\eta^2}{me}n^2. \quad (3)$$

It can be shown that the maximum value of $\langle d \rangle$ is four times more than in the classical case, and its probability distribution function, in contrast to the classical case, is constant.

3. The calculation of the spatial structure of the excited gas using the average dipole moment

The atomic configuration in this case determines not only by the spatial coordinates $\mathbf{r}_i$ but also by the value ($d_{\max}\sigma_i - 1 \leq \sigma_i \leq 1$) and direction $\mathbf{\Omega}_i$ of the dipole moment of each atom. The magnitude σ_i as well as the direction of vectors $\mathbf{\Omega}_i$ are equally probable. Using dimensionless coordinates $\boldsymbol{\rho}_i = N^{1/3}\mathbf{r}_i$ we introduce the interaction potential between atoms of different levels of excitation in the form of

$$V_{ij} = d_{\max}^2 \sigma_i \sigma_j (\mathbf{\Omega}_i \mathbf{\Omega}_j - 3(\mathbf{\Omega}_i \mathbf{k}_{ij})(\mathbf{\Omega}_j \mathbf{k}_{ij})/|\boldsymbol{\rho}_i - \boldsymbol{\rho}_j|^3), \quad (4)$$

limiting its strong repulsion at distances of a few Bohr radii. The exponent in the configuration integral (the weight function of Monte-Carlo calculations) has the form:

$$\exp(-\beta d_{\max}^2 \sum_{i < j} \sigma_i \sigma_j (\mathbf{\Omega}_i \mathbf{\Omega}_j - 3(\mathbf{\Omega}_i \mathbf{k}_{ij})(\mathbf{\Omega}_j \mathbf{k}_{ij})/|\boldsymbol{\rho}_i - \boldsymbol{\rho}_j|^3), \quad \mathbf{k}_{ij} = (\mathbf{r}_i - \mathbf{r}_j)/|\mathbf{r}_i - \mathbf{r}_j|. \quad (5)$$

When implementing the Monte-Carlo method, it is necessary to add to the usual change in spatial coordinates the steps with changes of magnitudes of σ_i and directions of vectors $\mathbf{\Omega}_i$. In

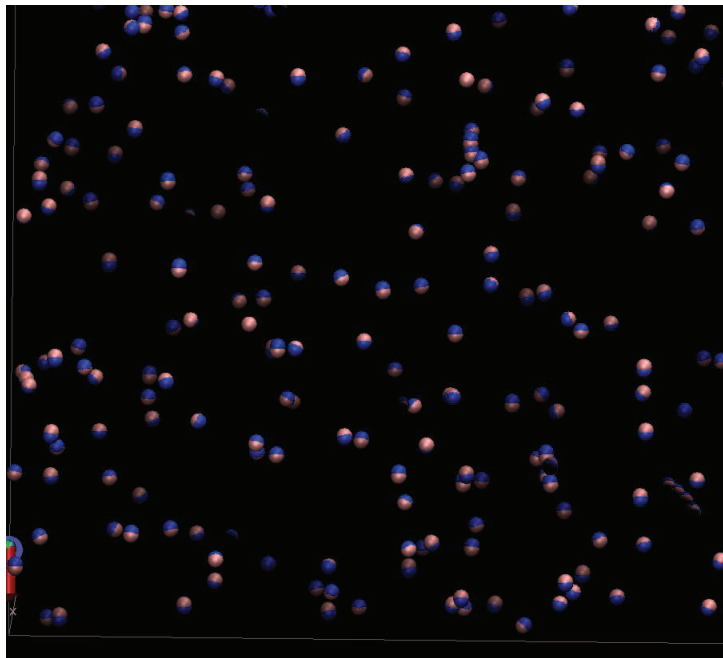


Figure 1. An example of spatial correlations of the dipoles for $\lambda = 0.075$. Light and dark color of the semispheres indicates the poles of the dipole.

order to determine the possible region of the phase transition it is necessary to estimate the nonideality parameter. The nonideality parameter can be determined as follows:

$$\lambda = N\beta d_{\max}^2 \approx \frac{Nn^2}{T} 10^{-24}, \quad (6)$$

where $[N] = 1 \text{ m}^{-3}$, $[T] = 1 \text{ K}$.

When $N = 10^{10} \text{ cm}^{-3} = 10^{16} \text{ m}^{-3}$, $T = 10^{-4} \text{ K}$, $n = 100$, one has $\lambda = 1$.

We have done calculations for different nonideality parameters.

Figures 1 and 2 show the typical configuration of an ensemble of particles in the periodic cell for different values of the nonideality parameter. The figures show that in case of greater value of λ the atoms put together in long chains.

4. Accounting for paired quantum effects

Let us consider the mixture of N_S atoms in S -state and N_P atoms in the P -state taking into account only the dipole interaction of atoms in different states. The configuration integral has the form:

$$Q = \int \prod_{i=1}^{N_S} d\mathbf{r}_i \prod_{j=1}^{N_P} d\mathbf{r}_j \Phi(\mathbf{r}_i, \mathbf{r}_j), \quad (7)$$

$$\Phi = \langle \Psi(\mathbf{r}_i, \mathbf{r}_j) | \exp(-\beta \sum_{i=1}^{N_S} \sum_{j=1}^{N_P} \hat{V}_{i,j}) | \Psi(\mathbf{r}_i, \mathbf{r}_j) \rangle, \quad (8)$$

$$\langle \Psi(\mathbf{r}_i, \mathbf{r}_j) | = \prod_i \langle S, \mathbf{r}_i | \prod_j \langle P, \mathbf{r}_j |, \quad (9)$$

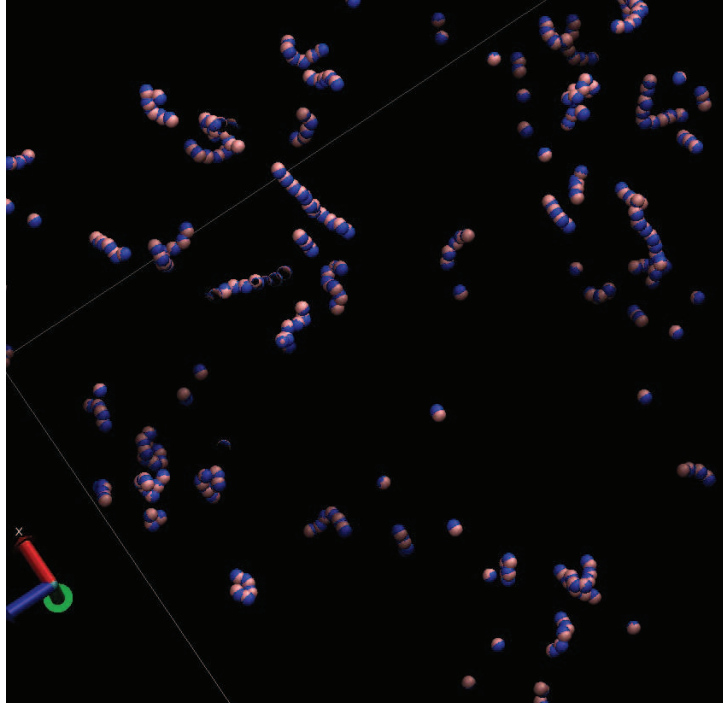


Figure 2. The only difference from the condition of figure 1 is $\lambda = 0.085$.

where $\langle S, \mathbf{r}_i |$ and $\langle P, \mathbf{r}_j |$ correspond to atoms in S -state at the point $\mathbf{r}_i$ in the P -state at a point $\mathbf{r}_j$:

$$\hat{V}_{i,j} = \frac{(\hat{\mathbf{d}}_i \hat{\mathbf{d}}_j) - 3(\hat{\mathbf{d}}_i \mathbf{k}_{i,j})(\hat{\mathbf{d}}_j \mathbf{k}_{i,j})}{|\mathbf{r}_i - \mathbf{r}_j|^3}. \quad (10)$$

In the pair approximation, with taking into account the quantum effects only in two-particle interactions, we have:

$$\Phi = \prod_{i=1}^{N_S} \prod_{j=1}^{N_P} \langle S, \mathbf{r}_i | \langle P, \mathbf{r}_j | \exp(-\beta \hat{V}_{i,j}) | S, \mathbf{r}_i \rangle | P, \mathbf{r}_j \rangle. \quad (11)$$

Replacing each factor in the last formula by symmetric one does not change the value of the configuration integral, but makes it easy to calculate the value of this multiplier. In this case note that P -states are characterized by a value of m (the projection of orbital momentum on z -axis). By averaging on m , we get finally for the weight function the expression:

$$\Phi = \prod_{i=1}^{N_S} \prod_{j=1}^{N_P} (2 \cosh(\beta d^2/3 |\mathbf{r}_i - \mathbf{r}_j|^3) + \cosh(2\beta d^2/3 |\mathbf{r}_i - \mathbf{r}_j|^3)) \vartheta(|\mathbf{r}_i - \mathbf{r}_j| - a)/3, \quad (12)$$

a is the hard core diameter, d is independent on m the reduced matrix element of the operator $\hat{\mathbf{d}}$. For large R , this value of each factor behaves like $1 + \beta^2 d^4/9R^6$ ($R = |\mathbf{r}_i - \mathbf{r}_j|$), and when R is small as $\exp(2\beta d^2/3R^3)/6$. The latter implies that the calculations should not allow the approach of the particles at the distance of about the size a for atom. To do this, we added a multiplier $\vartheta(|\mathbf{r}_i - \mathbf{r}_j| - a)$. When the concentration of particles $n \approx 10^{12} \text{ cm}^{-3}$ the argument in $\cosh$ changes to 12 orders of magnitude when R changes from a_0 to the average interatomic distance.

For the evaluation of the resonance transition in Li ($n = 2$) one can use the expression of the known lifetime of the P -states ($\tau \approx 0.3 \times 10^{-7}$ s) with dipole moment:

$$\frac{1}{\tau} = \frac{1}{4\pi\epsilon_0} \frac{4\omega^3 d^2}{3\hbar c^3} \quad (13)$$

($\omega \approx 2.8 \times 10^{15}$ s $^{-1}$).

For the maximum depth of the potential well at the point $R = a$ ($a = a_0 = \hbar^2/mc^2$) we have:

$$\frac{2d^2}{3a_0^3 Ry} = \frac{1}{8\pi\epsilon_0} \frac{c^3 m^2 e^2}{\omega^3 \hbar^3 \tau} \approx 3. \quad (14)$$

Apparently the value ~ 0.1 is more realistic that means a larger radius of the solid sphere $a = 3a_0$.

5. Conclusion

Some physical models are formulated in the paper to calculate the thermodynamics of the gas of Rydberg atoms. We obtained the spatial correlation of the dipoles in the equilibrium state for different nonideality parameters. It is established that there is a formation of chains of dipoles under certain nonideality parameters.

Acknowledgments

This work was supported by the Russian Science Foundation (grant No.14-19-01492).

References

- [1] Malnev V N and Pekar S I 1966 *Sov. Phys. JETP* **51** 1811
- [2] Malnev V N and Pekar S I 1970 *Sov. Phys. JETP* **58** 1113
- [3] Anderson W R, Veale J R and Gallagher T F 1998 *Phys. Rev. Lett.* **80** 249
- [4] Mourachko I *et al* 1998 *Phys. Rev. Lett.* **80** 253
- [5] van Ditzhuijzen C S E, Koenderink A F, Hernandez J V, Robicheaux F, Noordam L D and van Linden van den Heuvell H B 2008 *Phys. Rev. Lett.* **100** 243201
- [6] Zelener B B *et al* 2014 *J. Exp. Theor. Phys.* **100** 408
- [7] Zelener B B *et al* 2015 *J. Exp. Theor. Phys.* **148** 1086
- [8] Landau L D and Lifschitz E M 1959 *Quantum Mechanics* (Oxford: Pergamon Press)