Metastable superpositions of ortho- and para-Helium states

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Abstract

We analyze superpositions of ortho- and para-Helium states, considering the possible existence of stationary and metastable states in the system. In particular, the metastable superposition of $1s2s$ ortho and para states seems to be accessible to experimental scrutiny.

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1 Introduction

One of the most interesting manifestations of the principle of antisymmetrization of two-fermion systems is the existence of two types of configurations for the Helium atom. They correspond to singlet and triplet states and are usually denoted as para- and ortho-Helium states, showing different distributions of energy levels.

The study of the Helium atom is not a closed subject [1]. For instance, calculations of some energy levels of the confined Helium atom [2] and refinements in the para- and ortho-Helium evaluations [3] have been presented in the literature. In has also been shown that the ionization properties of the Helium atom are strongly dependent on the type of configuration [4]. The antiprotonic Helium [5], the system produced by the capture of an antiproton by a He^+ ion, has also extensively been studied.

In this Letter we suggest a new line to study the interesting properties of para- and ortho-Helium. We shall show that it is possible to prepare the Helium atom in a metastable superposition of para and ortho states. The scheme of preparation is in principle very simple. It is based on the capture by an He^+ ion of an electron, which must be prepared in a superposition of spin states. As we shall see later, in order to obtain a superposition one must only demand to the Hamiltonian describing the capture process to be symmetric. We should stress that this scheme appears quite naturally during the double ionization of He with intense laser fields. It has been shown that, for laser intensities below 5×10^{15} W/cm^2, the relevant path for ionization is a non-sequential process, in which the two electrons entangled are emitted with the same direction [6]. Since the probability for single ionization is always higher than the double, there is also a fraction of He^+ so the electron capture is also feasible. Note however that this process involves three particles, therefore the final superposition of He states is entangled with the surviving electron. Up to our knowledge, superpositions of ortho and para states have only been considered in the context of Helium collisions [7] (see section 3).

The proposed states would be interesting in several aspects. From a fundamental point of view they would provide us with a situation where the linearity of quantum mechanics has not been explored before, that in which the antisymmetrization postulate and exchange effects must be taken into account. Superpositions of different energy states of an atom have previously been considered in the literature. An atom can be prepared in a superposition of excited energy states using an impulsive excitation such that its Fourier spectrum contains frequency components corresponding to the energy intervals between ground and excited states. The rate of spontaneous emission of atoms prepared in this manner can oscillate in time. These quantum beats differ from the usual exponential decay expected for atoms in well-defined energy states [8]. Quantum beats, manifested as modulations in the absorption rates, are also present in the absorption of light by atoms in superposition states [9]. However, in these (energy-type) superpositions the exclusion principle does not play any role.

From a more practical point of view, it has been discussed the existence of collisional velocity changes associated with atoms in superposition states [10]. It has also been signaled that atomic superposition states are sensitive to phase dependent properties of radiation fields and, consequently, could be employed as detectors [11]. Moreover, we can expect that many properties of the atom such as absorption rates, ionization energy , etc. will differ in normal and superposition states and could be useful to understand the mechanisms involved in these processes.

Before considering these possibilities we must analyze the stability of the states of the superposition. In general, as we shall see in Sect. 3, there are only stationary states in some particular cases, when the ortho and para states are degenerate or almost degenerate. In the absence of stationary states we must consider the existence of metastable states, which could be studied with high resolution laser spectroscopy techniques. We shall study the superposition of ortho- and para-1s2s states, showing that it can be prepared as a long lifetime metastable superposition state. Moreover, in principle, this state is accessible to interesting experimental verifications such as the modification of the amplitude of the quantum beats and the variation of the mean lifetime and of the fluctuations of the decaying rate. In this context it must be signaled that the metastable $1s2s$ ortho-Helium state has been Bose condensed [12].

2 Preparation of the superposition

First of all we show how to prepare the superposition. On the one hand, we must have an He^+ ion with the electron in a well-defined state of the spin component along a given axis, for instance $|\uparrow>_z$. We take as axis of reference the z one, denoting by $|\uparrow>_z$ and $|\downarrow>_z$ the two possible states. We can determine that the electron is in the correct state by a direct measurement or by preparation. In the last case we should have an He^{++} ion, which captures an electron in the state $|\uparrow>_z$.

On the other hand, once prepared the He^+ ion with the electron in the $|\uparrow>_z$ state, we must have an electron in a superposition state $|\phi>=\alpha|\uparrow>_z +\beta|\downarrow>_z$, with $|\alpha|^2+|\beta|^2=1$. This step can be easily done by preparing the electron in a well-defined state along other spin axis. For instance, the up and down components of the spin along the two axes orthogonal to the z one are $(|\uparrow>_z +|\downarrow>_z)/\sqrt{2}$ and $(|\uparrow>_z -i|\downarrow>_z)/\sqrt{2}$. An alternative procedure, as discussed above, is to consider the non-sequential double ionization of He. Now we can make interact the electron in state $|\phi>$ with the He^+ ion, whose state is described by the ket $|He^+>$. If He^+ would only interact with an electron in state $|\uparrow>_z$, capturing it, the map describing the interaction would be

$$|He^+>|\uparrow>_z\rightarrow|He_{or}> \tag{1}$$

We would obtain an ortho-Helium atom because we have assumed that the other electron was also in the state $|\uparrow>_z$. On the other hand, if the ion He^+ would interact with an electron in the state $|\downarrow>_z$, the evolution would be

$$|He^+>|\downarrow>_z\rightarrow|He_{pa}> \tag{2}$$

In this case we would obtain a para-Helium state. Finally we move to the most interesting situation, that with the incident electron in a superposition of spin states. If the capture of the electron by the ion is a linear process (we discuss this point later) we have

$$|He^+>|\phi>\rightarrow\alpha|He_{or}>+\beta|He_{pa}> \tag{3}$$

which is a superposition of the Helium atom in para and ortho states.

We note that there is no superselection rule preventing the superpositions considered here. From all the superselection rules presented so far in the literature the only one that is related to our proposal is that preventing the existence of superpositions with different values of $(-1)^{2J}$, representing J the modulus of the total angular momentum, $\mathbf{J}=\mathbf{L}+\mathbf{S}$, with $\mathbf{L}$ the orbital angular momentum and $\mathbf{S}$ the spin. The two states of the superposition must have the same value of $(-1)^{2J}$. In other words, $2J$ must be in both cases an even or an odd integer. This is so in our case because the values of l are $0, 1, 2...$ and $s = 1/2$. Then $2J = 2l + 1$ that is always odd.

We discuss now the linearity of the capture process. The Hamiltonian describing the capture must take into account two different types of interactions: (i)The electromagnetic, spin-orbit, spin-spin, etc. usual interactions in the atom. (ii)The exchange effects associated with the antisymmetrization of the wavefunction of two identical electrons. With respect to (i) it is well-known the linearity of the associated Hamiltonian. We consider now (ii). The wavefunction of the complete system (nucleus plus the two electrons) must be antisymmetrized with respect to the variables of the two electrons, $\Psi(\mathbf{x}, s_x; \mathbf{y}, s_y; \mathbf{Z}; t) = \psi(\mathbf{x}, s_x; \mathbf{y}, s_y; \mathbf{Z}; t) - \psi(\mathbf{y}, s_y; \mathbf{x}, s_x; \mathbf{Z}; t)$ where $\mathbf{x}$ and $\mathbf{y}$ are the spatial coordinates of the two identical particles, s_x and s_y refer to the spin components and $\mathbf{Z}$ includes all the variables related to the nucleus. It is simple to show adding the Schrödinger equations ruling the evolution of each ψ that Ψ only obeys a linear Schrödinger's equation when $\hat{H}(\mathbf{x}, s_x; \mathbf{y}, s_y; \mathbf{Z}; t) = \hat{H}(\mathbf{y}, s_y; \mathbf{x}, s_x; \mathbf{Z}; t)$ ($\hat{H}$ is the Hamiltonian of the system), that is, when the Hamiltonian is symmetric. Since all the Hamiltonians used in atomic physics fulfill this condition we must expect the capture process to be linear.

Finally, we briefly discuss the possibility of actually implementing the scheme suggested here. We start with a sample of He atoms, which is illuminated by a laser tuned in the adequate frequency to induce double ionization. Using electric fields (which do not modify the spins) we can separate the He^{++} ions from He atoms and He^{+} ions. Then a beam of electrons in the $|\uparrow>_z$ state interacts with the sample of He^{++} ions. The $He^{+}(|\uparrow>_z)$ ions produced by the capture of one electron are separated, using again an electric field, from the He^{++} ions and $He(|\uparrow>_z, |\uparrow>_z)$ atoms. Finally, a beam of electrons, for instance in the $|\uparrow>_x$ state, interacts with the sample of $He^{+}(|\uparrow>_z)$ ions. The ions that capture electrons become in a superposition state. Using once more an electric field we can separate them from the $He^{+}(|\uparrow>_z)$ ions. The beams of electrons can be obtained from sources producing them in arbitrary random spin states using Stern-Gerlach devices with adequate orientations. An interesting alternative is provided by the process of non-sequential double ionization of He in strong electromagnetic process. In this case, the initial state is an entangled electron pair ionized from some neighbor atom. The capture of one of the electrons by a He^{+} ion, leads to a entangled state of the Helium atom with the surviving electron. The properties of such three particle system will be the subject of a future investigation.

3 Stationary states of the superposition

A fundamental question to be answered about the superposition is its stability. In the quantum realm an atom can be stable because of the existence of stationary states. We must look for the stationary states of the superposition, which would be given by the solutions of the equation

$$\hat{H}(\alpha|He_{or}[n]> +\beta|He_{pa}[m]>) = E_{[n,m]}(\alpha|He_{or}[n]> +\beta|He_{pa}[m]>) \quad (4)$$

where $[n]$ and $[m]$ represent the two sets of indexes characterizing the stationary states of both types of configurations. As $\hat{H}|He_{or}[n]>= E_{[n]}|He_{or}[n]>$ and $\hat{H}|He_{pa}[n]>= E_{[m]}|He_{pa}[m]>$ the stationary states of the superposition must obey the relation

$$E_{[n,m]} = E_{[n]} = E_{[m]} \quad (5)$$

In general, the energies of ortho and para states are different. But still it can happen that although with different energies, the states be so close to be considered as almost degenerate. As we shall discuss later in this section these states will decay by spontaneous emission. Then their energies will have an uncertainty determined by their mean lifetimes. In this context the natural criterion to consider two states as almost degenerate is that the difference between their energies be smaller than the broadening of the lines. As the mean lifetimes are of the order $10^{-9}s$, the uncertainty of the energies are $\delta E \approx 10^{-6}eV$ (note that this value is of the same order of the actual precision on the measurement of the energy). To see this point in detail let us analyze the experimental data. By the matter of concreteness we use the data of the NIST [13]. It is simple to see that there are some states for which the condition in Eq. (5) is fulfilled up to the broadening of the lines (and the experimental error). For instance, in the configuration $1s4f$ with terms $^3F^0$, $J = 2$ for the ortho and $^1F^0$, $J = 3$ for the para we have an energy difference between both levels $\Delta E = 9\,10^{-7}eV$, which is below δE (and the experimental error). Similarly, we have the configuration $1s5f$ with terms $^3F^0$,$J = 2$ and $^1F^0$,$J = 3$, or other terms obeying the relation $\Delta E \leq \delta E$.

Other degeneracy has already been signaled in the literature for this system. For $d3$ configurations the terms $2P$ and $2H$ are degenerate due to the symmetries existent in the problem.

We conclude that there are some energy levels of the ortho- and para-Helium which can be considered as almost degenerate. However, these states associated with almost degenerate levels would be of no interest since they would be unstable because of spontaneous emission. For instance, in the case of the state $1s4f$ the electron in the state $4f$ would emit photons decaying consecutively to states $3d$, $2p$ (final state in the ortho case) or $1s$ (final state for the para configuration). In the absence of stationary states of interest the most relevant states of the system would be the metastable ones. We consider then in next section.

We must signal here that F levels are involved in the only (up to our knowledge) previous consideration of mixed multiplicity states (see [7] and references therein). They are states that cannot be considered as pure singlets or triplets ones but rather as mixtures of them. They are related to the F-cascade model, in which it is assumed that in a collision process the excitation energy is transferred from resonance (n^1P)-levels to mixed F-levels: $He(1^1S) + He(n^1P) \rightarrow He(n^{mix}F) + He(1^1S)$, where $n^{mix}F$ represents a superposition of n^1F and n^3F states.

4 Metastable states

In the Helium atom the $1s2s$ state is metastable for both para- (lifetime of $19.7ms$) and ortho-type (lifetime around 10^8s) configurations. In the first case the decaying occurs through a two-photon electric dipole transition and in the second via relativistic and spin-orbit interactions. Consequently, we expect their superposition also to be a metastable state. We shall explore its properties.

First of all, we note that the energies of the two configurations are different, $E(1s2s, or) = E_{or} \neq E_{pa} = E(1s2s, pa)$. Then in addition to the superposition of the spins we must have a small indetermination in the energy of the state (initially carried by the incident electron), giving rise also to a superposition of the energy states. Both superpositions are compatible because energy and spin are compatible variables, not affected by complementarity relations.

Next we consider the lifetime of the state. As usual, the lifetime of a state is evaluated as the inverse of its decay rate, $\tau = 1/\Gamma$. If we denote by $|\phi_{or}>= |1s2s, or>$, $|\phi_{pa}>= |1s2s, pa>$ and $|\psi>= |1s1s, pa>$ (the ground state, at which both decay) the decay rate is given by

$$\Gamma = |<\psi|\hat{H}_*|\alpha\phi_{or} + \beta\phi_{pa}>|^2 = |\alpha M_{or} e^{-iE_{or}t/\hbar} + \alpha M_{pa} e^{-iE_{pa}t/\hbar}|^2 = \\ |\alpha|^2\Gamma_{or} + |\beta|^2\Gamma_{pa} + 2Re(\alpha^*\beta M_{or}^* M_{pa} e^{-i(E_{pa}-E_{or})t/\hbar}) \quad (6)$$

where $M_i =<\psi|\hat{H}_*|\phi_i>$, $i = or, pa$, are the matrix elements giving the transition rates, $\Gamma_i = |M_i|^2$. Note that we use $\hat{H}_*$ instead of $\hat{H}$ to remark that now relativistic and spin-orbit interactions are included.

Γ is the instantaneous value of the decay rate. However, it is unobservable. We must consider the averaged value $\overline{\Gamma}$ over the time scale T characteristic of the observations, $\overline{\Gamma} = 1/T\int_0^T \Gamma dt$. Assuming by simplicity the coefficients α and β and the matrix elements to be real we have for the interference term $\Gamma_{\alpha\beta}/T\int_0^T cos(\omega t)dt = \Gamma_{\alpha\beta} sin(\omega T)/\omega T$ where $\Gamma_{\alpha\beta} = 2\alpha\beta M_{or}M_{pa}$ and $\omega = (E_{pa} - E_{or})/\hbar$. As $\omega \approx 10^{15}s^{-1}$ and $|sin(\omega T)| \leq 1$ we have that $sin(\omega T)/\omega T \approx 0$ for any T much larger than $10^{-15}s$. Consequently, for any realistic T the interference term can be neglected in the averaged expressions. Finally, we can write

$$\tau = \frac{1}{|\alpha|^2\Gamma_{or} + |\beta|^2\Gamma_{pa}} \quad (7)$$

The lifetime of the superposition state ranges between $\tau_{or} = 1/\Gamma_{or}$ and $\tau_{pa} = 1/\Gamma_{pa}$. Varying the coefficients α and β we can obtain all the values of lifetimes in that range. For any value of the coefficients we have a metastable state.

Although the interference term has not influence on the (mean) lifetimes its effects lead to high frequency oscillations of the decay rate, in a similar way to the quantum beats present in energy-type superposition states. To emphasize this point we follow the approach of Ref. [8] expressing the coefficients αM_{or} as $A_{or}e^{-\Gamma_{or}t}$,..., i. e., taking into account explicitly the dependence of the coefficients on the decay rate. Hence, the probability for the transition at time t (when all the amplitudes are assumed to be real valued) is $A_{or}^2 e^{-2\Gamma_{or}t} + A_{pa}^2 e^{-2\Gamma_{pa}t} + 2A_{or}A_{pa}e^{-(\Gamma_{or}+\Gamma_{pa})t} cos(\omega t)$, showing clearly the existence of oscillations around the mean values. These oscillations, being their amplitude and frequency of the same order of magnitude than those associated with energy-type superpositions, are in principle experimentally observable. Moreover, varying the parameters α and β we could modulate the amplitude of the oscillations.

Another experimental way to test the existence of the superpositions would be the measurement of the variations of the decaying rate. We proceed in the standard way, i. e., by counting the number of decays in a given time interval of observation. From an experimental point of view the way of measuring the decays is to count the photons emitted during that interval. As different atoms emit their radiation independently this photon source is of chaotic type. As it is well known [14] the distribution of photocounts is of Poisson type, $P_n(T) = <n>^n \; e^{-<n>}/n!$, provided that the time of observation T is much longer than the coherence time of the light (if not the distribution would be super-Poisson). In the above relation $<n>$ denotes the mean number of photocounts, which in the semiclassical approximation (for chaotic light the semiclassical and fully quantum approaches give the same result [14]) is given by the expression $<n> = \xi \overline{I} T$, with ξ the efficiency of the detectors and $\overline{I} = \overline{I}(t)$ the cycle-averaged intensity of the light. The intensity is given by the number of photons emitted at t. By definition, this number is proportional to the number of atoms that decay at t, which is Γ, and $I(t) \sim \Gamma(t)$. Finally, we must average over the time of observation, which must be much longer than the coherence time of the light. Denoting by $\overline{\Gamma}$ this average over T (which according to our previous results is time-independent for any realistic choice of T) the mean number of photocounts is $<n> = \overline{\Gamma} T$, where the efficiency factor has been absorbed in $\overline{\Gamma}$ by simplicity in the notation. The Poisson distribution can be expressed as

$$P_n(T) = \frac{(\overline{\Gamma} T)^n}{n!} exp(-\overline{\Gamma} T) \tag{8}$$

Using the relation $\overline{\Gamma} = |\alpha|^2 \Gamma_{or} + |\beta|^2 \Gamma_{pa}$ and the well-known expression $(x+y)^n = \sum x^{n_x} y^{n_y} n!/n_x! n_y!$, where the summation extends to all the non-negative integers obeying the relation $n_x + n_y = n$, we have,

$$P_n(T) = \sum_{n_{or}+n_{pa}=n} \mathcal{P}_{n_{or}}(T)\mathcal{P}_{n_{pa}}(T) \tag{9}$$

where

$$\mathcal{P}_n(T) = \frac{(|\alpha|^2\Gamma_{or}T)^{n_{or}}}{n_{or}!} exp(-|\alpha|^2\Gamma_{or}T) \tag{10}$$

that is, the same (8) distribution, but with the decay rate replaced by $|\alpha|^2\Gamma_{or}$, a weighted decay rate. Therefore, the detection distributions show a characteristic dependence on α and β that could be tested experimentally. Moreover, the distribution (9) differs from that expected for a mixture of Helium atoms prepared in $(1s2s)$ ortho and para states with weights $|\alpha|^2$ and $|\beta|^2$, which as it is simple to see is given by the expression $P_n^{mix}(T) = \sum_{n_{or}+n_{pa}=n} |\alpha|^2 P_{n_{or}}^{or}(T)|\beta|^2 P_{n_{pa}}^{pa}(T)$, with $P_{n_{or}}^{or}$ and $P_{n_{pa}}^{pa}$ the usual Poisson distributions with Γ_{or} and Γ_{pa}.

We conclude that, in principle, some peculiar characteristics of the metastable $1s2s$ superposition state can be observed experimentally.

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