# Decay Kinetics and Bose Condensation in a Gas of Spin-Polarized Triplet Helium

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We consider the decay kinetics of a trapped spin-polarized gas of metastable triplet helium $^4$He($2\,^3S$) at ultralow temperatures. The sample lifetime is found to be determined by spin relaxation and Penning ionization, both induced by spin-dipole interaction in pair collisions. The rates of these processes are calculated. The Penning ionization proves to be 5 orders of magnitude slower than in the unpolarized case. The results indicate that spin-polarized triplet helium is a promising candidate for Bose-Einstein condensation.

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The prospect of observing macroscopic quantum phenomena, such a Bose-Einstein condensation (BEC), in trapped atomic gases generates a great deal of interest, especially in view of recent successes in cooling magnetically trapped hydrogen [1,2] and alkali atoms [3,4] to microkelvin temperatures. Metastable triplet helium, helium in the $2\,^3S$ state (He*), is an attractive candidate for such investigations. He* is a unique example of a long-lived excited atomic state, with a radiative lifetime $\tau_0 \approx 8 \times 10^3$ s [5,6]. It can be magnetically trapped and, with a Doppler limit for laser cooling of 40 $\mu$K, and a recoil limit of 2 $\mu$K, is well suited for optical manipulation with the LNA laser [7]. Furthermore, subrecoil cooling mechanisms are being developed [8].

Is it possible to create a low-temperature gaseous phase of He* with sufficient density to observe BEC? The He* research performed thus far, including experiments in cryogenic environments [9] and magneto-optical traps [10], shows He* as a very unstable gas. The dominant decay mechanism of the unpolarized gas, Penning ionization

$$\text{He}(2\,^3S) + \text{He}(2\,^3S) \rightarrow \begin{cases} \text{He}^+ + \text{He}(1S) + e\,, \\ \text{He}_2^+ + e \end{cases} \quad (1)$$
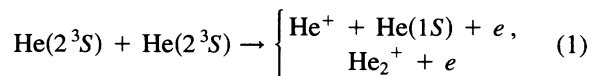
is so fast (rate constant $\sim 10^9$ cm$^3$/s [11]) that it easily dominates over elastic collisions at low temperatures. It is well known that spin polarization should lead to an essential reduction in the Penning ionization rate as the total spin of the colliding particles in the final state does not exceed 1, whereas in the initial state it equals 2, and the spin conservation rule is not satisfied. However, a suppression of the ionization rate of polarized atoms by more than 1 order of magnitude as compared to the unpolarized case has not been established [9].

In this Letter we present a theoretical analysis of the decay kinetics of spin-polarized $^4$He* ($^4$He*↑) at ultralow temperatures and show that full spin polarization should lead to suppression of the Penning ionization rate of 5 orders of magnitude. This offers the opportunity to create a gaseous phase of $^4$He*↑ of sufficient stability to enable evaporative and optical cooling in magnetostatic traps

and to reach the densities and ultralow temperatures relevant for BEC. Magnetostatic trapping automatically provides us with spin polarization. Loading of a deep superconducting trap to high density can, in principle, be accomplished from a cryogenic discharge, with the $^4$He*↑ thermalized, before reaching the surrounding walls, by diffusion in ground-state He vapor [12].

The critical temperature for Bose condensation is related to the gas density $n$ by $T_c = 3.31\hbar^2 n^{2/3}/m$, where $m$ is the atom mass. For $^4$He*↑ and $n = 10^{14}$ cm$^{-3}$ we have $T_c \sim 10\ \mu$K. Two decay mechanisms, both induced by the spin-dipole interaction in pair collisions, limit the achievable density of $^4$He*↑. The first one is spin relaxation resulting in escape of atoms from the trap. This process was very well investigated in particular for hydrogen [13,14]. An important difference originates from the absence of hyperfine interaction in the case of $^4$He. This leads to a strongly field dependent relaxation rate, becoming vanishingly small in very low fields. A fundamental lower bound on the decay rate of $^4$He*↑ in low fields is set by relaxation-induced Penning ionization: a mechanism in which virtual spin-dipole transitions to the zero spin state of the quasimolecule lift the spin-conservation rule and induce ionization through the ordinary Penning mechanism (cf. [15]).

A principal question with regard to obtaining Bose condensates in atomic gases concerns the sign of the elastic scattering length $a$. If $a > 0$, the effective interaction between atoms is repulsive and the condensate will be stable with respect to elastic interaction. If $a < 0$, the effective interaction is attractive and should cause a collapse of the condensate. Using the $^5\Sigma_g^+$ potential calculated in [16] we find a scattering length for $^4$He*↑ which is large and positive and remains positive under variation of the potential within its quoted accuracy. However, a firm prediction of the value and even the sign of $a$ usually requires additional information [17]. We return at the end of this Letter to the importance of a large positive scattering length for the problem of Bose condensation in trapped gases.

In this Letter we confine ourselves to inelastic collisional processes in $^4$He*↑ under the condition

$$kR_e \ll 1 \qquad (2)$$

($s$-wave scattering limit), where $k = \sqrt{mT}/\hbar$ is the thermal wave vector and $R_e$ is the characteristic radius of elastic interaction. The $^5\Sigma_g^+$ potential $U_2(R)$ of interaction between two $^4$He*↑ atoms is characterized by a fairly deep potential well ($\sim$1500 K [16,18]) containing many bound states [the $^1\Sigma_g^+$ interaction potential $U_0(R)$ in the zero spin state of the quasimolecule has an even deeper well $\sim$8000 K [18]]. A recent calculation of $U_2(R)$ [16] results in the highest $s$ level having a binding energy of several millikelvin. Our analysis shows that $R_e$ is close to the value $70a_0$ ($a_0$ is the Bohr radius) obtained from the condition $\hbar^2/mR_e^2 = |U_2(R_e)| \equiv E_e$. Hence, condition (2) corresponds to $T \ll E_e = 10$ mK.

In the case of $^4$He*↑, having spin 1, the spin-dipole interaction in pair collisions

$$\hat{H}_{\text{int}} = \frac{4\mu_B^2}{R^5} [(\hat{S}_1 \cdot \hat{S}_2)R^2 - 3(\hat{S}_1 \cdot \mathbf{R})(\hat{S}_2 \cdot \mathbf{R})] \qquad (3)$$

($\hat{S}_1$, $\hat{S}_2$ are the spin operators of the colliding atoms and $\mathbf{R}$ is the internuclear distance) can change not only the spin projection $M$ of the quasimolecule, but also the total spin $S$. Representing the spin wave function of the initial state of the quasimolecule ($S = 2, M = 2$) as $\Phi_{22} = \Phi_1^{(1)}\Phi_1^{(2)}$, one can see that $\hat{H}_{\text{int}}$ causes transitions to the following final spin states $\Phi_{SM}$:

$$\Phi_{21} = \left[\Phi_1^{(1)}\Phi_0^{(2)} + \Phi_0^{(1)}\Phi_1^{(2)}\right] / \sqrt{2},$$

$$\Phi_{20} = \left[\Phi_1^{(1)}\Phi_{-1}^{(2)} + \Phi_{-1}^{(1)}\Phi_1^{(2)} + 2\Phi_0^{(1)}\Phi_0^{(2)}\right] / \sqrt{6}, \qquad (4)$$

$$\Phi_{00} = \left[\Phi_1^{(1)}\Phi_{-1}^{(2)} + \Phi_{-1}^{(1)}\Phi_1^{(2)} - \Phi_0^{(1)}\Phi_0^{(2)}\right] / \sqrt{3}.$$

Here $\Phi_{M_1}^{(1)}$ and $\Phi_{M_2}^{(2)}$ are the spin states of the colliding spin-1 atoms, with spin projections $M_1$ and $M_2$, respectively.

Transitions to states with $S = 2$ correspond only to spin relaxation. Atoms with opposite or zero spin projection on the direction of the magnetic field, produced in the relaxation process, either escape from the trap or enter the Penning process (1). Importantly, the transitions to the $S = 0$ state $^1\Sigma_g^+$, which is autoionizing via the ordinary Penning mechanism, involve "relaxation-induced" ionization as well as spin relaxation.

As the spin-dipole interaction is much weaker than the elastic interaction between particles, the transition rates for relaxation and relaxation-induced ionization can be calculated within first-order perturbation theory.

(a) When the change of the Zeeman energy in the transition, $E_M = 2\mu_B B(2 - M) \gg T$, the transitions predominantly occur at distances $R \ll 1/k$. Then the initial state wave function of the relative motion of atoms in the potential $U_2(R)$ may be represented by the zero energy $s$-wave contribution $\sqrt{2}\,\xi(R)$, where $\xi \to 1$ for $R \to \infty$; the factor $\sqrt{2}$ is due to identity of nuclei. Because of the

nonspherical character of $\hat{H}_{\text{int}}$ (3), the scattered wave of atoms for each spin state (4) will only contain the partial $d$-wave $Y_{2,2-M}(\mathbf{R}/R)\phi_{SM}(R)$, with

$$\phi_{SM}(R) = \frac{\sqrt{2}\,m}{\hbar^2 R} \int G_{SM}(R,R')H_{SM}(R')\xi(R')R'\,dR'. \qquad (5)$$

Here $G_{SM}(R,R')$ is the Green function of the Schrödinger equation for the radial motion in the potential $U_S(R)$ with orbital angular momentum $l = 2$ and energy $E_M$. At $R = R'$, $G_{SM}(R,R')$ is continuous and the discontinuity in the derivative $\partial G_{SM}(R,R')/\partial R$ is equal to 1. The quantity $H_{SM}(R) = (384\pi A_{SM}/5)^{1/2}\mu_B^2/R^3$ comes from the transition matrix element over the spin variables. The coefficients $A_{SM}$ take the following values: $A_{21} = 1$, $A_{20} = 2/3$, and $A_{00} = 1/3$. For $R \to \infty$ we have

$$G_{SM}(R,R') = \frac{\chi_S(k_f,R')}{2k_f} \exp(ik_f R), \qquad (6)$$

where the momentum of the scattered wave $\hbar k_f = \sqrt{mE_M}$. The function $\chi_S(k_f,R)$ describes elastic $d$ scattering of an incident plane wave $\exp(i\mathbf{k}_f \cdot \mathbf{R})$ in the potential $U_S(R)$.

When calculating $\chi_0(k_f,R)$ one should account for the Penning ionization which occurs with probability close to unity at $R \lesssim 7a_0$ (see [18]). As the spin-dipole transitions occur at larger $R$, we use a simple model: We put a perfectly absorbing boundary at a distance $R_0 \simeq 7a_0$ and consider the potential to be purely elastic and equal to $U_0(R)$ at larger $R$. Then we have a boundary condition $\chi_0(k_f,R) \sim \exp[-ik_{f0}(R - R_0)]$ at $R \to R_0$, with $k_{f0} = \hbar^{-1}\{m[E_M - U_0(R_0) - 6\hbar^2/mR_0^2]\}^{1/2}$. The precise value of $R_0$ is not important because the radial motion at $R \sim R_0$ is quasiclassical.

For $R \to R_0$ the Green function $G_{00}(R,R')$ takes the form

$$G_{00}(R,R') = \frac{\tilde{\chi}_0(k_f,R')}{2(k_f k_{f0})^{1/2}} \exp[-ik_{f0}(R - R_0)], \qquad (7)$$

where the function $\tilde{\chi}_0(k_f,R)$ describes elastic scattering of an outgoing spherical $d$ wave, "starting" at $R = R_0$, by the potential $U_0(R)$. For $R \to R_0$ this function contains the outgoing spherical wave and its reflection from the centrifugal barrier. For $R \to \infty$, $\tilde{\chi}_S(k_f,R) = \sqrt{1 - |S_0(k_f)|^2}\,\exp(ik_f R)$, where $S_0(k_f)$ is the $S$-matrix element for elastic $d$ scattering of an incident plane wave.

The spin relaxation rates are determined from the radial flux of particles in the scattered wave $\phi_{SM}(R)$ at $R \to \infty$. Outside the BEC regime, representing the event rate per unit volume as

$$\nu_{\text{rel}} \equiv \alpha_{\text{rel}}n^2/2 = (\alpha_{21} + \alpha_{20} + \alpha_{00})n^2/2, \qquad (8)$$

for the relaxation rate constant $\alpha_{SM}$ in each relaxation channel (4) from Eqs. (5) and (6) we obtain

$$\alpha_{SM} = A_{SM}\alpha_*(E_M/E_e)^{1/2}F_S(E_M), \qquad (9)$$

where $\alpha_* = (512\pi/15)(\mu_B^2 m/\hbar)^2(E_e/m)^{1/2} \simeq 1.3 \times 10^{-14}$ cm$^3$/s and

$$F_S(E_M) = \left| \frac{3}{2k_f} \int_0^\infty \xi(R)\chi_S(k_f, R)\frac{dR}{R^2} \right|^2 . \quad (10)$$

The same result is obtained in the distorted-wave approximation.

In low magnetic fields, where $E_M \ll E_e$ (but still $E_M \gg T$), the main contribution to the integral comes from $R \sim 1/k_f \gg R_e$. Then we can take $\xi(R) = 1$ and use the wave function of free motion for $\chi_S(k_f, R)$ in Eq. (10). Also for $S = 0$ this is allowed because Penning ionization can be neglected for such $k_F$ [$|S_0(k_f)| \simeq 1$]. This gives $F_S = 1$ and, hence, $\alpha_{SM} \sim \sqrt{B}$. Actually, for large positive scattering length $a$, which is the case for the potential $U_2(R)$ [16], one should use $\xi(R) = 1 - a/R$. Then for fields where $k_f a \sim 1$ there is a strong cancellation of the contributions of the two terms in $\xi(R)$, and the field dependence of $\alpha_{SM}$ should show a dip (see also Fig. 1). Observation of this dip would firmly establish the sign of $a$.

In higher fields, where $E_M \gtrsim E_e$, the functions $\xi(R)$ and $\chi_2(k_f, R)$ and the function $\chi_0(k_f, R)$ were found numerically for the potentials $U_2(R)$ [16] and $U_0(R)$ [18], respectively. The rate constants $\alpha_{SM}$ reach their maximum values in the fields 400–800 G ($E_M \sim E_e$). Further increase of $B$ leads to decreasing relaxation rate, with $\alpha_{00}$ decreasing more rapidly than $\alpha_{21}$ and $\alpha_{20}$. For low $B$ ($E_M \ll E_e$) the functions $F_S(E_M) \simeq 1$ and we recover the result of the free motion approximation. The field dependence of $\alpha_{\text{rel}}$, corresponding to Eq. (8), in the zero temperature limit is presented in Fig. 1.

The rate constant of relaxation-induced ionization is determined from the radial flux of atoms in the scattered wave $\phi_{00}(R)$ onto the absorbing boundary at $R_0$. Representing the event rate per unit volume due to relaxation-induced ionization by $\nu_{\text{ri}} = \alpha_{\text{ri}}n^2/2$, from Eqs. (5) and (7) we arrive for the rate constant of this process, $\alpha_{\text{ri}}$, at
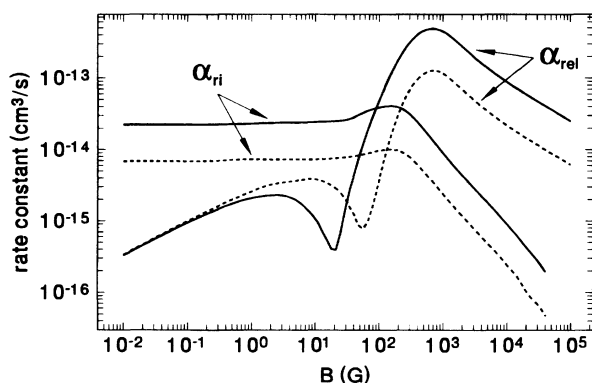


FIG. 1. Rate constants for spin relaxation ($\alpha_{\text{rel}}$) and relaxation-induced Penning ionization ($\alpha_{\text{ri}}$). Solid curves correspond to the potential $U_2(R)$ [16], and dashed curves to the same potential multiplied by 1.01.

Eqs. (9) and (10) with $S = M = 0$ and $\chi_0(k_f, R)$ replaced by $\tilde{\chi}_0(k_f, R)$.

The wave function $\tilde{\chi}_0(k_f, R)$ was found numerically for the potential $U_0(R)$ [18]. The field dependence of $\alpha_{\text{ri}}$ in the zero temperature limit is shown in Fig. 1. In fields $B \lesssim 100$ G it is field independent ($\alpha_{\text{ri}} \sim 10^{-14}$ cm$^3$/s) and dominates over spin relaxation. In high fields, where $E_M$ greatly exceeds the height of the centrifugal barrier ($E_M \gg E_e$), the radial motion is quasiclassical at any $R$ and we have $\tilde{\chi}_0(k_f, R) = \chi_0^*(k_f, R)$. Hence, $\alpha_{\text{ri}} \approx \alpha_{00}$, i.e., rapidly decreases with increasing $B$ and becomes smaller than $\alpha_{\text{rel}}$.

Our results are rather sensitive to the potential $U_2(R)$ because it supports a weakly bound $s$ level. The accuracy of $U_2(R)$ is discussed in detail in [16]. Possible improvement of this potential is likely to make it deeper (not more than 1%) and increase the binding energy of this $s$ level, which will decrease $\alpha_{\text{rel}}$ and $\alpha_{\text{ri}}$. As an example, we also present in Fig. 1 the results of calculations using the potential $1.01U_2(R)$.

(b) In very low magnetic fields, where $E_M \lesssim T \ll E_e$ and $k_f$ is comparable with $k$, spin relaxation is temperature dependent. In this case, due to the $1/R^3$ dependence of the spin-dipole interaction, the relaxation transitions occur at distances $R \sim 1/k$ and the scattered wave can contain many partial waves. Using the plane wave approximation and assuming a Boltzmann distribution, we obtain

$$\alpha_{SM} \approx A_{SM}\alpha_* \left( \frac{T + 4\pi E_M}{4\pi E_e} \right)^{1/2} . \quad (11)$$

For $E_M \gg T$ Eq. (11) reduces to Eq. (9) with $F_S = 1$.

Irrespective of the ratio between $k_f$ and $k$, the main contribution to the probability of relaxation-induced ionization comes from distances $R \lesssim R_e$. Hence, the zero energy limit for $\alpha_{\text{ri}}$ is valid at any ratio between $E_M$ and $T$, as long as the condition (2) is satisfied.

Another ionization channel in pair collisions of $^4$He*↑ atoms is Penning ionization via the direct dipole-exchange mechanism [15]. The probability of dipole-exchange ionization at a fixed $R$ relates to the probability of ordinary Penning ionization as the fourth power of the fine structure constant. For $T \ll 10$ mK we estimate the rate constant of dipole-exchange ionization to be $\alpha_i \sim 10^{-16}$ cm$^3$/s, independent of magnetic field and temperature. In any realistic $B$ this process is slower than either relaxation-induced ionization or relaxation.

Spin relaxation and Penning ionization in $^4$He*↑ may also be induced by spin-orbit interaction. We have estimated that the corresponding rates are significantly smaller than those due to spin-dipole interaction.

In $^4$He*↑ also three-body recombination will be present. As there are bound states in the potential well $U_2(R)$, recombination can occur due to elastic interaction between $^4$He*↑ atoms in the course of three-body collisions and

lead to the formation of a spin-polarized $He_2^*$ molecule decaying through Penning ionization due to spin-dipole interaction. The estimate of the recombination rate constant $\alpha_{rec}$ on the basis of the characteristic radius of interaction, $R_e$, gives $\alpha_{rec} \lesssim 10^{-30}$ cm$^6$/s. Despite the existence of a weakly bound $s$ level in the potential $U_2(R)$, which increases $\alpha_{rec}$ by approximately 2 orders of magnitude, the recombination rate $\alpha_{rec}n^3$ is expected to be smaller than $\nu_{ri}$ for achievable densities ($n \lesssim 10^{14}$ cm$^{-3}$) of $^4$He$^*\uparrow$.

The results obtained in this Letter indicate the feasibility of achieving BEC in $^4$He$^*\uparrow$. The main decay channels are connected with spin relaxation and Penning ionization in pair collisions of atoms due to their spin-dipole interaction. Spin relaxation proves to be strongly field dependent, and is substantially reduced in low fields. For $B \lesssim$ 100 G the characteristic decay time $\tau$ of the gas is determined by relaxation-induced ionization, $\tau \sim 1/\alpha_{ri}n$, and should be of order seconds or larger for $n \lesssim 10^{14}$ cm$^{-3}$ corresponding to Bose condensation at temperature $T \lesssim$ 10 $\mu$K. In high fields the dominant decay channel is spin relaxation. The rate constant $\alpha_{rel}$ decreases with increasing $B$, but does not drop below the low-field value of relaxation-induced ionization even at the highest fields investigated. Therefore low fields are more promising for achieving BEC.

An important circumstance making $^4$He$^*\uparrow$ attractive for BEC is a large possible value of the elastic cross section $\sigma$. With $R_e \simeq 70a_0$ we expect $\sigma \sim 10^{-12}$ cm$^2$, i.e., 3 orders of magnitude larger than in spin-polarized hydrogen. This has an important consequence for evaporative cooling and the formation kinetics of a Bose condensate. Both are determined by the rate constant of elastic collisions, $\alpha_{el} = \sigma\langle v \rangle \sim \sqrt{T}$. In $^4$He$^*\uparrow$ we expect the ratio $\alpha_{ri}/\alpha_{el}$ to be 2 orders of magnitude smaller than the corresponding quantity $\alpha_{rel}/\alpha_{el}$ in hydrogen. For evaporative cooling of $^4$He$^*\uparrow$ this implies the possibility to reach BEC at much lower temperatures, maybe even in the nanokelvin regime. With a large positive scattering length, the effective elastic interaction $4\pi\hbar^2 a/m$ will be repulsive and much stronger than in hydrogen, reducing the compression of the condensate by the trapping field. This diminishes the enhancement of the inelastic processes in the condensate and allows a deeper penetration into the BEC regime (cf. [19]).

In common with experiments on any trapped gas, with $^4$He$^*\uparrow$ care must be taken to avoid excessive heating of the sample by the inelastic processes. Heating occurs because inelastic processes preferentially occur low in the trap, where the density is highest, and because the reaction products may collide with trapped atoms before escaping,

and should be compensated by the mechanisms employed to cool the gas. We do not foresee any debilitating heating in the case of trapped $^4$He$^*\uparrow$.

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