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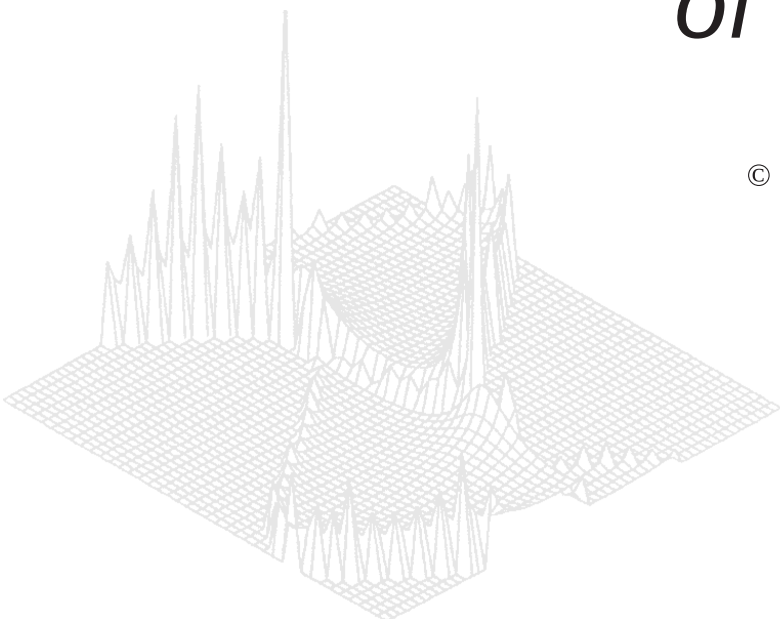

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A Monte Carlo Study of Radiation Trapping Effects*

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Abstract

A Monte Carlo simulation of radiative transfer in an atomic beam is carried out to investigate the effects of radiation trapping on electron–atom collision experiments. The collisionally excited atom is represented by a simple electric dipole, for which the emission intensity distribution is well known. The spatial distribution, frequency and free path of this and the sequential dipoles were determined by a computer random generator according to the probabilities given by quantum theory. By altering the atomic number density at the target site, the pressure dependence of the observed atomic lifetime, the angular intensity distribution and polarisation of the radiation field is studied.

1. Introduction

It is well known that the radiation field emitted by an excited atom is, in general, polarised and anisotropic. By measuring the polarisation and angular distribution of the emitted photons, one can retrieve information on the configuration of the excited atomic states, generally represented by a density matrix $\rho = \sum f_{m'} f^*_m |\varphi_{m'}\rangle\langle\varphi_m|$. Here f_m are the quantum mechanical complex amplitudes, describing the population of and the coherence between each atomic eigenstate φ_m. These amplitudes reflect the interactions and dynamics of the associated excitation process (Andersen *et al*. 1988). The study of electron–atom collisions has recently been pursued with the aim of obtaining a complete knowledge of these complex amplitudes. The atomic state of interest is selected by detecting the scattered energy-loss electron in time coincidence with the photon emitted by the excited atom. In this way, a particular radiation pattern is associated with a single electron–atom collision event.

However, a measurement is always performed with finite atomic number density at the target site, resulting in a finite probability of absorption and re-emission of radiation by the neighbouring atoms especially between the excited and the ground states. In particular, when the associated excitation cross section is low, the target density is usually increased until sufficient collisions occur within a practical time scale to give statistically acceptable signal rates. Because the re-emitted

* Dedicated to Professor Robert W. Crompton on the occasion of his seventieth birthday.

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photons still correlate in time with the scattered electron, they contribute to the coincidence signal. The consequence is that the apparent time taken for the detection of radiation is prolonged and it is usually called radiation trapping. This process can also change the polarisation and angular intensity distribution of the radiation field. As a result, the measured radiation field would present a false image of the excited atom especially when the target density is high.

Williams *et al.* (1992), Williams and Wang (1994) and Mikosza *et al.* (1994) studied the effects of radiation trapping on the measured lifetime and the Stokes parameters of collisionally excited atoms. Their analysis was based on solutions of the Boltzmann equation in the absence of boundary conditions. The difficulty in solving the Boltzmann equation with the characteristic dimensions of an apparatus motivated the present study. Instead of solving the complicated equation, a computer simulation of the radiative transfer process for atomic helium was performed. By altering the atomic number density in the gas cell, the pressure dependence of the measured angular intensity distribution and polarisation of the radiation field was studied.

2. Formulation of the Problem

The Monte Carlo method has been applied to simulate the radiation transfer process by several researchers. For example, Hishikawa *et al.* (1992) studied the influence of radiation trapping on the measured linear polarisation of the helium $501\cdot 6$ nm line in collision spectroscopy. We adopted a similar approach for the study of the radiation trapping effects on angular distributions of the radiation field and lifetimes measured at various angles.

The sequence of the radiation trapping process is as follows. At time $t = 0$, a helium atom is excited to the 2^1P state and is represented by a simple dipole. After the characteristic lifetime of the excited atom, it decays back to the ground state by emitting a photon at a certain emission angle. This photon travels a certain distance (on average the mean free path) through the gas cell until it collides with another helium atom in its ground state. This atom then absorbs the photon and is excited. After a certain time the second excited atom decays by emitting another photon. Such photon emission and absorption processes continue until a photon reaches the detector or is lost to the walls of the apparatus.

Four stochastic elements are involved in the above process. The decay time is sampled at random according to the probability distribution

$$P(t) = \gamma e^{-\gamma t}\,, \tag{1}$$

where $\gamma = 1/\tau$ and τ is the natural lifetime of the excited state. For the 2^1P state of helium, $\tau = 0\cdot 56 \pm 0\cdot 02$ ns. The probability distribution function for the emission angle is

$$P(\theta,\varphi) = \tfrac{3}{4}\sin^3\theta\,, \tag{2}$$

where θ and φ are the polar and azimuthal angles, respectively. Due to the thermal motion of the atoms, the Doppler effect causes a Gaussian spread in the frequency of the emitted photons with a probability function,

$$P(\nu) = \frac{1}{\sqrt{\pi}}\left(\frac{c}{v_0\,\nu_0}\right)\exp\left[-\left(\frac{c}{v_0}\right)^2\left(\frac{\nu-\nu_0}{\nu}\right)^2\right], \tag{3}$$

where c is the speed of light, $v_0 = \sqrt{2RT/m}$ is the thermal velocity of the atom, R is the Boltzmann constant, T is the temperature, m is the mass of the atom, and ν_0 is the resonance frequency. Finally, the probability distribution function for the photon's free path x is

$$P(x) = \exp[-a(\nu)x]\,, \tag{4}$$

where the absorption coefficient* is

$$a(\nu) = \frac{1}{8\pi\sqrt{\pi}}\left(\frac{Nc^3\gamma}{v_0\,\nu_0^3}\right)\left(\frac{g_2}{g_1}\right)\exp\left[-\left(\frac{c}{v_0}\right)^2\left(\frac{\nu-\nu_0}{\nu}\right)^2\right]. \tag{5}$$

Here g_1 and g_2 are the statistical weights of the ground and excited states respectively, and N is the atomic number density of atoms existing in the ground state.

3. Random Number Generation

The generation of pseudo-random numbers is of great importance in a Monte Carlo simulation utilising a large number of deviates. The basis of generating non-uniform random deviates according to some probability distribution is a uniform random number generator that must be 'good' in the sense that it should be uniformly distributed, successive deviates should be statistically independent, and it should be reproducible. However, we also require speed and have memory restrictions on a computer so that a compromise between these factors is sought.

A large variety of uniform random number generators is available of which many fall into the class of congruential generators. These are based on the fundamental congruence relationship $x_{i+1} = (ax_i + c)(\text{mod}\, m), i = 1, ..., n$ and are the basis of internal random number generators in many digital computers. However, such generators have a period of m and are therefore cyclic which can introduce unwanted periodic effects into a simulation. The type of generator chosen for use depends largely on the number of deviates it is expected to produce.

Three different generators were tested, a simple type of congruential generator (Press *et al.* 1986), the Decstation 5000/2000 machine generator and a shuffled generator (Press and Teukolsky 1992). It is found that all three types of generator satisfy the chi-square tests for both distribution and independence, with the shuffled generator having the lowest χ^2 values but using slightly more CPU. The basis of this generator is that it takes two different uniform generators and shuffles their outputs to remove low order correlations between successive random

* A typographical error is noted in Hishikawa *et al.* (1992) for the same formula.

numbers. It is quoted to have a periodicity of $>10^8$ deviates (Press and Teukolsky 1992), while the present simulation program requires about 10^7 random numbers in total, including the decay time, the azimuthal emission angle, the emission frequency, and the free path. These variables are produced with different starting values to further reduce correlation effects. The shuffled generator is adopted in the present work. Fig. 1 shows the probability distribution of randomly generated numbers according to the above established functions for the decay time, the azimuthal emission angle, the emission frequency and the free path.

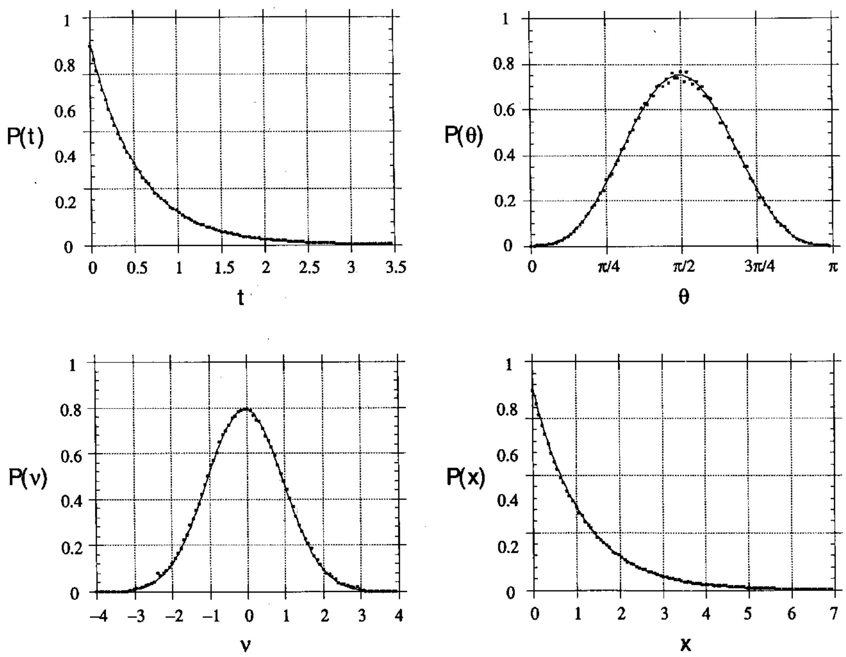

Fig. 1. Probability distribution for the decay time, the azimuthal emission angle, the emission frequency, and the free path. The solid curves show the expected distribution functions and the dotted lines represent the probability distributions of randomly generated numbers.

4. Results

The actual simulation consists of piecing together the various stochastic elements. It is assumed that the radiative transfer process is confined to a spherical region where the atoms are uniformly distributed. Once a photon leaves this 'gas sphere' it travels without further collision to a circular array of detectors placed at a large distance from the origin compared with the radius of the gas sphere. When a photon finally emerges from the gas sphere and hits a detector, one count is registered for that particular detector. Also recorded are the total travel time and the time taken for the intermediate excited atoms to decay. The simulation is run at a number of different pressures while all other factors are held constant. In this way, we studied the pressure dependence of the angular distribution of the radiation field and the measured atomic decay rates.

(4a) Angular Distribution of the Detected Photons

For each pressure, the simulation was run until a total of $0 \cdot 5$ million photons were registered over all detectors. The results of these calculations are plotted in Fig. 2, which shows the angular distribution of the emitted photons for three different pressures. When there are effectively no neighbouring atoms (i.e. at extremely low pressure), the simulated results agree with the expected angular intensity distribution of a simple electric dipole. Fig. 2 shows that at a pressure of $2 \cdot 0$ mTorr a definite broadening of the angular distribution has occurred. As the pressure is increased further the aspect ratio of the intensity distribution is seen to decrease. In other words, the polarisation of the observed radiation field changes as the pressure in the gas cell changes. A similar broadening was measured and reported in Fig. 8 of Mikosza *et al.* (1994).

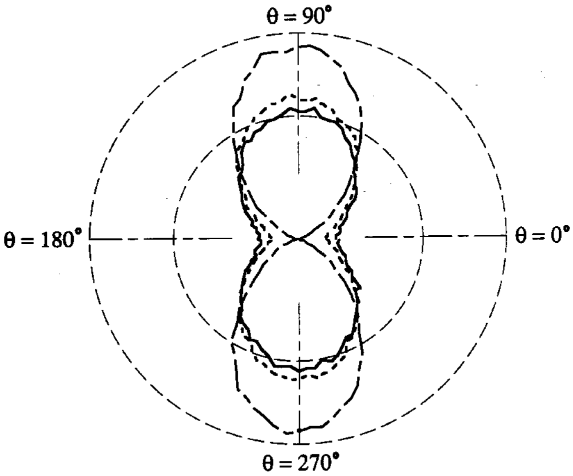

Fig. 2. Angular intensity distribution of the detected photons from the simulation for zero pressure (— – —), $2 \cdot 0$ mTorr (– – –) and $3 \cdot 15$ mTorr (——).

According to equation (2), the probability of a photon being emitted at an angle θ near $0°$ or $180°$ is close to zero. This means that very few photons are detected at such emission angles if the radiation trapping process does not occur (i.e. at extremely low pressure). However, at a pressure high enough for several radiative transfers to occur before a given photon is detected, we see in Fig. 2 that a significant number of photons are recorded at these angles, and a broadening of the radiation pattern occurs. When a photon is absorbed, the resultant dipole is perpendicular to the direction of the incoming photon and thus the dominant angle of emission is rotated from that of the preceding dipole. As the pressure increases, the mean free path of the photon decreases and the photon may undergo more radiative transfers before escaping the gas cell. As a result, there is an increased expectation that one of the induced dipoles will emit a photon near $\theta = 0°$ or $180°$. This is essentially what we see in Fig. 3 where the locus of a particular photon trajectory follows a 'curved' path resulting in a finite probability for photons detected at angles of $0°$ or $180°$.

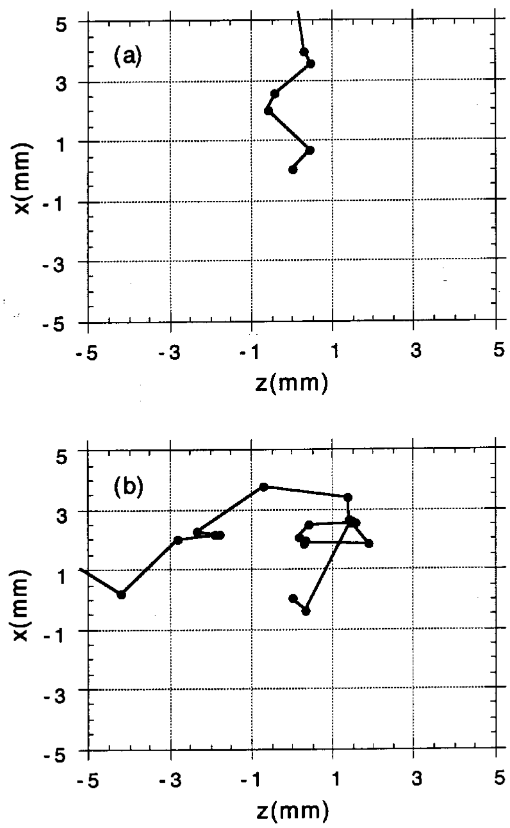

Fig. 3. Typical trajectories of radiation transfer projected onto the xz plane at a pressure of $3\cdot 15$ mTorr. The final photon is detected near (a) $\theta = 90°$ and (b) $\theta = 180°$.

(4b) Time Spectra of Detected Photons

The more interesting result in Fig. 3 is the apparently different number of steps taken for a typical photon to arrive at $\theta = 90°$ and $\theta = 180°$, although the pressure at the target site is the same for both cases. This indicates that the measured lifetime can be significantly different if the photon detector is positioned at different angles to the original dipole. To study this further, we plot in Fig. 4a the time spectra recorded at various positions and in Fig. 4b the extracted values for the lifetime. It is clearly shown that the observed lifetime has increased by more than 10% by viewing the time spectrum at 0° compared with at 90° at a pressure of $6\cdot 2$ mTorr. The higher the pressure, the larger the difference. A qualitative explanation for this effect is that at higher pressure the paths of more photons will be redirected by the radiation trapping process towards 0° or 180° and these photons statistically undergo more radiative transfers than photons detected at 90° as shown in Fig. 3. The consequence is that the average time taken for the photons to emerge from the system is longer for the detector at 0° in comparison with that at 90°. This phenomenon has also been observed in a real system as shown in Fig. 6 of Williams and Wang (1994).

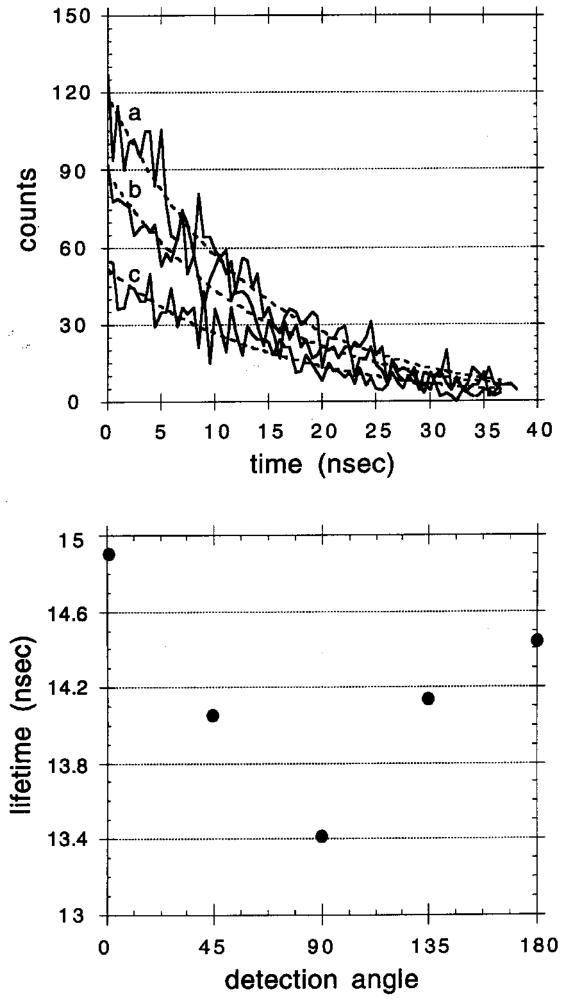

Fig. 4. Upper panel: time spectra recorded at three detection angles for a fixed pressure of 6·2 mTorr. The detectors are located at (*a*) 90°; (*b*) 45° and (*c*) 0°. Lower panel: lifetimes extracted by fitting exponential functions to the measured time spectra.

The most significant effect of radiation trapping is the strong pressure dependence of the observed lifetime. Fig. 5*a* shows the time spectra measured at 90° for various pressures at the target site. Fig. 5*b* shows the extracted lifetimes from these spectra. From this plot we can see the large effect that radiation trapping has on the observed lifetime of an atomic state. The observed lifetime has increased by a factor of approximately 36 when the pressure reaches about 7·1 mTorr. Note that at zero pressure the simulated lifetime is 0·555 ns which agrees well with the lifetime of 0·56 ns used as an input parameter in the simulation code. This apparent increase in lifetime is due to a photon having a mean free path dependent on the pressure. As the pressure is increased, the mean free

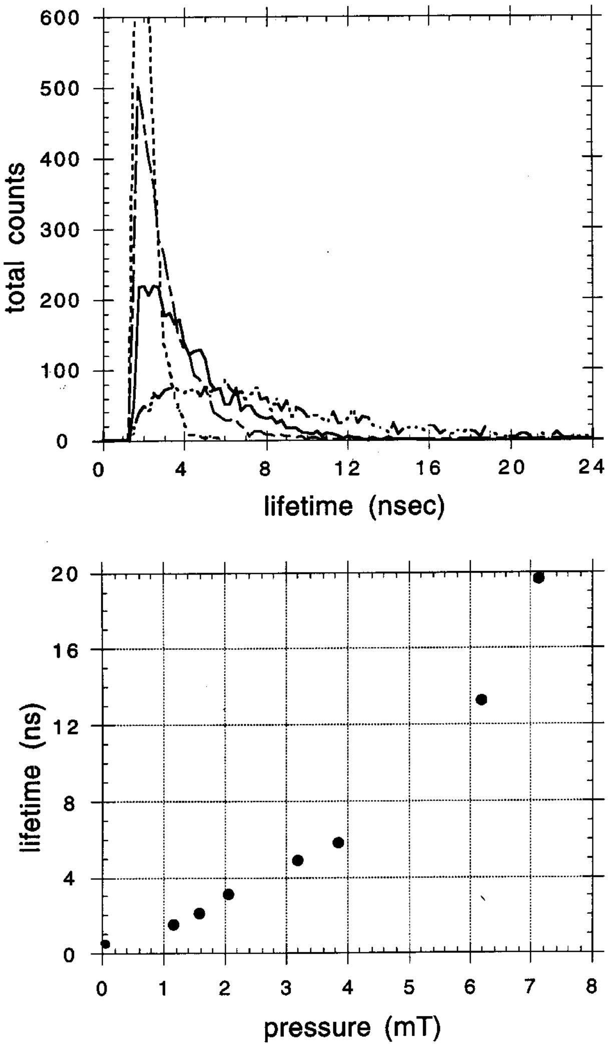

Fig. 5. Upper panel: time spectra recorded at four different pressures: zero (– – –), 1·1 mTorr (— – —), 2·0 mTorr (——) and 3·8 mTorr (— – - – —). Lower panel: lifetimes extracted by fitting exponential functions to the measured time spectra.

path of a photon decreases; hence to reach a detector a photon will have to undergo more radiative transfers. Due to the finite lifetime of each dipole the more times a photon is transferred the longer its emergence time will be. A statistical estimate of the average number of radiative transfers per photon at a particular pressure can be made by dividing the observed lifetime by the natural lifetime, i.e. for a pressure of 7·1 mTorr approximately 36 transfers occur before a photon reaches the detector.

5. Conclusion

From a qualitative viewpoint, the present simulation has successfully produced the radiation trapping effects as observed in experiments, namely the prolonged lifetime as target site pressure increases, the pressure-broadening of radiation intensity distribution, and the different lifetimes observed at different angles. Further work will be performed so as to make quantitative comparisons with experiment, where a more realistic model than the simple electric dipole will be used to represent the excited atom. It is hoped that this model can then be applied to determine accurately the pressure at the target site.

Acknowledgments

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