SLAC-PUB-6158
April 1993
T

Atomic Self-Coherence*

RICHARD BLANKENBECLER

Stanford Linear Accelerator Center

Stanford University, Stanford, California 94309

ABSTRACT

There is nothing new in this paper. It contains a pedagogical discussion of a very interesting interference effect involving the radiation pattern from a single atom which is confined by a multi-well potential. The interference phenomenum is a quantum effect with a pattern strength that is proportional to the tunneling probability. Extensions of the phenomenon are briefly discussed.

Submitted to *Journal of Physics A.*

* Work supported by the Department of Energy, contract DE–AC03–76SF00515.

1. Introduction

After viewing the images of individual atoms as provided by electron microscopes, especially when they have been arranged to spell out the initials of a rather well-known company[1], it is easy to slip into the error of thinking of atoms as classical billiard balls with quantum phenomena becoming important at a smaller distance scale—except for exceptional circumstances such as superconductivity. The purpose of this note is pedagogical; we will describe the familiar process of the radiation of a single photon by an atom. The unusual feature is that the radiation interferes with itself by virtue of the fact that the atom is in a non-localized state with a net symmetry. If the atom is in a multi-well potential, with a finite tunneling probability, the radiation pattern will in general contain structure that reflects the geometric arrangement of the wells. The magnitude of this variation is proportional to the quantum tunneling probability.

New laboratory techniques in the manipulation of individual atoms have opened up new possibilities in both experiment and theory. This paper discusses the interference pattern expected from a single atom bound in a multi-well potential is discussed. Suitable binding potentials should be available to realize this type of interference, e.g., in molecules with an appropriate structure[2], by selecting suitable defects on the surface of a solid[3], or by an appropriate configuration of external electromagnetic fields in a Stern-Gerlach interferometer[4].

To simplify our discussion and to permit an emphasis on the physics of the interference phenomena, we will consider the problem of radiative decay of an excited hydrogen-like (alkali) atom bound in a double well potential that is a

function of the center-of-mass coordinate only. The nonrelativistic Hamiltonian[5]

$$H = \frac{1}{2M_e}\,(p_e + eA(r_e))^2 + \frac{1}{2M_p}\,(p_p - eA(r_p))^2 + V(\vec{r}) + W(\vec{R})\,, \qquad (1.1)$$

where $\vec{r} = \vec{r}_e - \vec{r}_p$ and $\vec{R} = (M_e\vec{r}_e + M_p\vec{r}_p)/M_T$ (see Appendix B for a complete set of definitions). The potential $V(r)$ is the atomic binding potential; $W(R)$ is the center-of-mass potential that is assumed to have two well-separated minima at $\vec{R} \cong \pm\vec{D}/2$ in the example treated below.

The standard electromagnetic interaction is then given by the linear current interaction:

$$J \cdot A = \frac{e}{M_e}\,\vec{p}_e \cdot \overrightarrow{A}(r_e) - \frac{e}{M_p}\,\vec{p}_p \cdot \overrightarrow{A}(r_p)\,, \qquad (1.2)$$

which can be rewritten in relative coordinates (see Appendix B) as

$$J \cdot A = \frac{e}{M_r}\,\vec{p}_r \cdot \overrightarrow{A}(r_e) - \frac{e}{M_p}\,\vec{p}_p \cdot \left[\overrightarrow{A}\,(r_p) - \overrightarrow{A}\,(r_e)\right]\,. \qquad (1.3)$$

The last term can be safely neglected in the limit of large M_p and small k (the dipole approximation limit). Let us now turn to a discussion of photon emission from a free and then a bound atom.

2. Single Atom–Dipole Emission

As a warmup, and for later use, we will first consider the radiation of a single photon of momentum k from an atom in the dipole approximation for two different situations: a free atom and an atom bound in a center-of-mass potential. The interaction Hamiltonian is approximated as

$$H' = J \cdot A = \frac{e}{M_r}\, \vec{p}_r \cdot \overrightarrow{A}(r)\; e^{-i\mathbf{k}\cdot R} \qquad \Big\lfloor_{\to\, \vec{\epsilon}\, e^{-i\mathbf{k}\cdot r} \sim \vec{\epsilon}} \tag{2.1}$$

$$H' = \frac{e}{M_r}\, \vec{p}_r \cdot \vec{\epsilon}\, e^{-i\mathbf{k}\cdot R} \equiv v\, e^{-i\mathbf{k}\cdot R}\ , \tag{2.2}$$

where ϵ is the photon polarization vector, and standard manipulations lead to

$$v = \frac{e}{M}\, \vec{p}_r \cdot \vec{\epsilon} = \frac{e}{2M}\, [H, \vec{r}] \cdot \vec{\epsilon} \to \frac{e\,k}{2M}\, \vec{r} \cdot \vec{\epsilon} \to \ldots \text{ etc.} \tag{2.3}$$

First we will discuss the emission from a 'spinless' free atom and thereby choose the wave functions as:

$$\begin{aligned} \text{Initial State} \quad |i\rangle &= e^{iP\cdot R}\,|p\rangle \qquad |p\rangle = 2p \text{ state} \\ \text{Final State} \quad |f\rangle &= e^{iP'\cdot R}\,|s\rangle \qquad |s\rangle = 1s \text{ state} \end{aligned} \tag{2.4}$$

with energies E_p and E_s, respectively. The transition matrix element then becomes

$$M = \langle s\,|\,v\,|\,p\rangle \int e^{-i(P'+k\cdot P)\cdot R} d^3R = \langle s\,|\,v\,|\,p\rangle\, (2n)^3 \delta(\vec{P}' + \vec{k} - \vec{P})\ , \tag{2.5}$$

and the differential decay rate is written succinctly as

$$\text{Rate} \sim \sum |M|^2 \cong \sum_{pol} |\,\langle s\,|\,v\,|\,p\rangle\,|^2 \rho \equiv R_{\text{free}} \tag{2.6}$$

where $E_p - E_s = k_0 = |k|$, and recoil energy has been neglected.

If the atom is in a simple confining potential $W_S(R)$, in a state of energy E_S with well-separated energy levels, the wave functions for the transition in which only the state of the *relative* wave function changes can be written

$$\begin{aligned} &\text{Initial State} \qquad |i\rangle = \phi(R)\ |p\rangle \\ &\text{Final State} \qquad |f\rangle = \phi(R)\ |s\rangle\ , \end{aligned} \tag{2.7}$$

with energies given by $E_S + E_p$ and $E_S + E_s$, respectively. The transition matrix element for remaining in the state E_S while the internal state changes is then

$$M = \langle s\,|\,v\,|\,p\rangle \int d^3R\, e^{-i\mathbf{k}\cdot\mathbf{R}}\phi^2(R) \equiv \langle s\,|\,v\,|\,p\rangle\ F(k)\ , \tag{2.8}$$

with $E_p - E_s = k_0 = |k|$ as before. The decay rate is written as

$$\text{Rate} = R_{\text{free}}|F(k)|^2\ , \tag{2.9}$$

where $F(k)$ is the form factor for the center-of-mass (C–M) bound state, measuring its ability to absorb a momentum transfer of k.

3. One Atom–Double Well

Following the notation and level of approximation of the preceding discussion, let us now consider the case in which the atom is in a C–M double well potential. The initial state is assumed to be the lowest symmetric eigenstate in the well-separated double well and will be approximated as

$$\Psi_i = |+\rangle = N_+ \left[\phi\left(\vec{R} + \frac{1}{2}\vec{D}\right) + \phi\left(\vec{R} - \frac{1}{2}\vec{D}\right)\right] |p\rangle \ , \tag{3.1}$$

where the normalized functions $\phi(R)$ peak at $R = 0$. The energy of the state is $E_+ + E_p$ where $E_+ = E_R - TE_T/2$ (T is the exponentially behaved overlap factor defined below), and E_T is a positive reasonable number of order E_R which is easily computed given the form of the potential. For clarity, we write the above as

$$\Psi_i = |+\rangle = N_+ \left[\phi(R+) + \phi(R-)\right] |p\rangle \ . \tag{3.2}$$

The corresponding anti-symmetric (spatial) state is

$$|-\rangle = N_- \left[\phi(R+) - \phi(R-)\right] |p\rangle \ , \tag{3.3}$$

with energy $E_- + E_p$ where $E_- = E_R + TE_T/2$.

The normalization factors are:

$$\frac{1}{N_\pm^2} = 2[1 \pm T] \ , \quad \text{where} \quad T = \int d^3R \, \phi(R+)\phi(R-) \ . \tag{3.4}$$

For a gaussian trial function, the C–M wave function, tunneling factor T and form

factor $F(k)$ are

$$\phi(R) \;=\; N\,e^{-R^2/2\,a^2} \quad , \quad T \;= e^{-D^2/4a^2} \quad \text{and} \quad F(k) \;= e^{-a^2k^2/4}\,. \tag{3.5}$$

The decay process will lead to both symmetric and antisymmetric final states:

$$\begin{aligned} |+\rangle &= N_+\,[\phi(R+) + \phi(R-)]\,|s\rangle \\ |-\rangle &= N_-\,[\phi(R+) - \phi(R-)]\,|s\rangle \end{aligned} \tag{3.6}$$

with energies $E_\pm + E_s$, respectively. The transition matrix element to the symmetric final state is

$$\begin{aligned} \langle +|\,H'\,|\,\Psi_i\rangle &= N_+^2\,\langle s\,|\,v\,|\,p\rangle \int d^3R\,e^{-i\mathbf{k}\cdot\mathbf{R}}[\phi(R+) + \phi(R-)]^2 \\ &= N_+^2\,\langle s\,|\,v\,|\,p\rangle \left\{e^{i(1/2)\,\mathbf{k}\cdot\mathbf{D}}F(k) + e^{-i(1/2)\,\mathbf{k}\cdot\mathbf{D}}F(k) + 2J\right\}\,, \end{aligned} \tag{3.7}$$

where $k = E_+ + E_p - E_+ - E_s = E_p - E_s$, and

$$\begin{aligned} J &\equiv \int d^3R\,e^{-i\mathbf{k}\cdot\mathbf{R}}\phi(R+)\phi(R-) \;\propto\; \int d^3R\,e^{-i\mathbf{k}\cdot\mathbf{R}}\;e^{-(1/a^2)[R^2+(1/4)D^2]} \\ J &= F(k)\,T\,, \end{aligned} \tag{3.8}$$

for a gaussian wave function. Thus $|\mathbf{k}| = E_p - E_s$, as before, and

$$\langle +|H'|\,\Psi_i\rangle = \langle s\,|\,v\,|\,p\rangle\;F(k)\;\left(\frac{\cos\frac{D\cdot k}{2} + T}{1+T}\right)\,. \tag{3.9}$$

The transition matrix element to the antisymmetric state can be written

$$\begin{aligned} \langle -|H'|\,\Psi_i\rangle &= \langle s\,|\,v\,|\,p\rangle\;N_+N_-\left\{e^{i(1/2)D\cdot k_-}F(k_-) - e^{-i(1/2)k_-\cdot D}F(k_-)\right\} \\ &= \langle s\,|\,v\,|\,p\rangle\;F(k)\,i\left(\frac{\sin\frac{D\cdot k_-}{2}}{\sqrt{1-T^2}}\right)\,, \end{aligned} \tag{3.10}$$

where $k_- \;= E_+ + E_p - E_- - E_s \;= k - TE_T$, and the difference between k and k_- has been retained in the phase factor only.

Assuming that the detector sums the counts from photons of energy k and k_-, the total decay rate from the symmetric initial state is then

$$\text{Rate} \;=\; R_{\text{free}}|F(k)|^2\, A_+ \;, \tag{3.11}$$

where the angular modulation factor A_+ contains the interference terms of interest; it is given by

$$A_+ \;=\; \left\{ \left(\frac{c+T}{1+T} \right)^2 + \frac{s_-^2}{1-T^2} \right\} \;, \tag{3.12}$$

where for convenience we have introduced

$$\theta \;=\; \frac{1}{2} D\cdot k \qquad \text{and} \qquad \theta_- \;=\; \frac{1}{2} D\cdot k_- \;\cong\; \theta\,(1 - T\,\frac{E_T}{k}) \tag{3.13}$$

and defined

$$\begin{aligned} c \;&=\; \cos\theta \\ s_- \;&=\; \sin\theta_- \;\cong\; s - cT\,\frac{E_T}{k}\,\theta \;. \end{aligned} \tag{3.14}$$

Thus to first order in T we find

$$A_+ \;=\; 1 + 2T\left[c(1-c) - c\,s\,\frac{E_T}{k}\,\theta \right] \;. \tag{3.15}$$

In the limit where $E_T \ll k$, the pattern becomes[6]

$$A_+ \;=\; 1 + 2T\; c(1-c) \;, \tag{3.16}$$

4. Physical Interpretation

The matrix element leading to the symmetric final state,

$$\begin{aligned}\langle +|H'|\,\psi_i\rangle &= \cdots\left\{\int d^3R\, e^{-ik\cdot R}\,[\phi(R+)+\phi(R-)]^2\right\} \\ &= \cdots\left\{\int d^3R\, e^{-ik\cdot R}\,\left[\phi^2(R+)+\phi^2(R-)+2\phi(R+)\,\phi(R-)\right]\right\}\ ,\end{aligned} \tag{4.1}$$

has the expected peaks at $\pm D/2$ and a small (overlap) peak at $R = 0$. The anti-symmetric final state produces *no* peak at $R = 0$,

$$\begin{aligned}\langle -|H'|\,\psi_i\rangle &= \cdots\left\{\int d^3R\, e^{-ik\cdot R}\,[\phi(R+)-\phi(R-)]\,[\phi(R+)+\phi(R-)]\right\} \\ &= \cdots\left\{\int d^3R\, e^{-ik\cdot R}\,\left[\phi^2(R+)-\phi^2(R-)\right]\right\}\ .\end{aligned} \tag{4.2}$$

If there were no peak at $R = 0$ and if $E_T \ll k$, so that $T = 0$ and $s_- \cong s$, then the total rate has no angular dependence. Interference arises from the overlap peak at $R = 0$ which is coherent with the charge peaks at $R = \pm\frac{1}{2}\,D$ by virtue of tunneling.

5. Initial ODD state

Following the previous discussion, we write

$$\chi_i = N_-[\phi(R+)-\phi(R-)]\,|p\rangle\ , \tag{5.1}$$

and then with $k_+ \equiv k + TE_T$ and $\theta_+ \cong \theta\,(1 + T\,\frac{E_T}{k})$,

$$\begin{aligned}
\langle +|H'|\,\chi_i\rangle &= N_+ N_- \,\langle s\,|\,v\,|\,p\rangle \int d^3R\, e^{-ik_+\cdot R}[\phi(R+) + \phi(R-)]\,[\phi(R+) - \phi(R-)] \\
&= \langle s\,|\,v\,|\,p\rangle \; F(k)\, i\, \frac{\sin\theta_+}{\sqrt{1-T^2}} \qquad\qquad \text{and} \\
\langle -|H'|\,\chi_i\rangle &= N_-^2 \,\langle s\,|\,v\,|\,p\rangle \int d^3R\, e^{-ik\cdot R}[\phi(R+) - \phi(R-)]^2 \\
&= \langle s\,|\,v\,|\,p\rangle \; F(k) \; \left(\frac{\cos\theta - T}{1-T}\right) .
\end{aligned} \tag{5.2}$$

Thus the total rate for this initial state is

$$\text{Rate} \;=\; R_{\text{free}} |F(k)|^2 \; A_- \;, \tag{5.3}$$

with

$$A_- \;=\; \left\{ \left(\frac{c-T}{1-T}\right)^2 + \frac{s_+^2}{1-T^2} \right\} \;, \tag{5.4}$$

and to first order in T, we find

$$A_- \;=\; 1 - 2T \left[c(1-c) - c\,s\,\frac{E_T}{k}\,\theta \right] \;. \tag{5.5}$$

6. Extensions—Conclusions

If the initial state has probability p_+ to be in the symmetric state, and hence a probability $p_- = 1 - p_+$ to be in the antisymmetric state, the resultant first order angular dependence is

$$A \;=\; 1 - (p_+ - p_-)\,T\;\left[c(1-c) - c\,s\;\frac{E_T}{k}\,\theta\right]\;. \tag{6.1}$$

Thus if the initial state is a 50/50 mixture of the symmetric and antisymmetric wave functions, the first order terms in T will cancel, and there will be no angular dependence to first order in the total rate.

Thus the statement made in the abstract is now proven; there is nothing new in this paper.

APPENDIX A

Alternative Final States

If the energy splitting in the final states is zero, i.e. $E_T = 0$, then any *orthogonal* linear combination of the degenerate pair is as valid as any other. To illustrate this, consider the set:

$$\begin{aligned} |L\rangle &= \phi(R+)\,|s\rangle \\ |R\rangle &= \frac{1}{\sqrt{1-T^2}}\,[\phi(R-) - T\phi(R+)]\,|s\rangle \end{aligned} \tag{A.1}$$

where $|L\rangle$ has the atom in the left well wave function and $|R\rangle$ has the atom mostly in the right well wave (but orthogonality is retained). Then starting from an initial state with a given symmetry, the relevant matrix elements are:

$$|\Psi_i\rangle = \frac{1}{\sqrt{2}\sqrt{1 \pm T}}\,[\phi(R+) \pm \phi(R-)]\,|p\rangle \tag{A.2}$$

$$\langle L|H'|\,\Psi_i\rangle = \frac{1}{\sqrt{2}\sqrt{1 \pm T}}\,\langle s\,|\,v\,|\,p\rangle\; F(k)\,\left[e^{i\theta} + T\right] \tag{A.3}$$

$$\langle R|H'|\,\Psi_i\rangle = \frac{1}{\sqrt{2}}\,\frac{1}{\sqrt{1 \pm T}}\,\frac{1}{\sqrt{1-T^2}}\,\langle s\,|\,v\,|\,p\rangle\; F(k)\,\left[\pm T + e^{-i\theta} \mp T\,e^{i\theta} - T^2\right] \tag{A.4}$$

$$\text{Rate} = R_{\text{free}}|F(k)|^2\,A_{\pm} \tag{A.5}$$

$$A_{\pm} \;=\; \left\{\left(\frac{c \pm T}{1 \pm T}\right)^2 + \frac{s^2}{1-T^2}\right\}\,, \tag{A.6}$$

which is exactly the same result as before in this limit of $E_T = 0$.

APPENDIX B

Standard Coordinate Transformation

In the text we have used the standard coordinate transformations to relative and center-of-mass variables:

$$\begin{aligned} \vec{r} &= \vec{r}_e - \vec{r}_p & \vec{r}_e &= \vec{R} + \frac{M_p}{M_T}\,\vec{r} \\ \vec{R} &= \frac{1}{M_T}\,(M_e\vec{r}_e + M_p\vec{r}_p) \quad \Rightarrow & \vec{r}_p &= \vec{R} - \frac{M_e}{M_T}\,\vec{r} \end{aligned} \tag{B.1}$$

$$M_T = M_e + M_p \ .$$

Now use the fact that $\vec{p}_e \cdot \vec{r}_e + \vec{p}_p \cdot \vec{r}_p = \vec{p}_{\rm TOT} \cdot \vec{R} + \vec{p}_r \cdot \vec{r}$ to write:

$$\begin{aligned} \vec{p}_{\rm TOT} &= \vec{p}_p + \vec{p}_e & \vec{p}_e &= \vec{p}_r + \frac{M_e}{M_T}\,\vec{p}_{\rm TOT} \\ \vec{p}_r &= \frac{1}{M_T}\,(M_p\vec{p}_e - M_e\vec{p}_p) \quad \Rightarrow & \vec{p}_p &= \frac{M_p}{M_T}\,\vec{p}_{\rm TOT} - \vec{p}_r \ , \end{aligned} \tag{B.2}$$

and, of course,

$$\frac{1}{2M_e}\,p_e^2 + \frac{1}{2M_p}\,p_p^2 = \frac{1}{2M_r}\,p_r^2 + \frac{1}{2M_T}\,p_{\rm TOT}^2 \ , \tag{B.3}$$

where the reduced mass is given by

$$\frac{1}{M_r} \equiv \frac{1}{M_e} + \frac{1}{M_P} \ . \tag{B.4}$$

ACKNOWLEDGEMENTS

The author wishes to thank Claude Itzykson for introducing this problem and for interesting and helpful discussions. Discussions with Hans Evertz are also gratefully acknowledged.

REFERENCES

1. D. M. Eigler and E. K. Schweizer, Nature **344**, 524 (1990).
2. Molecules with adjacent indentical binding sites, for example.
3. A surface with adjacent empty sites, or other interesting voids.
4. J. Robert, Ch. Miniatura, O. Gorceix, S. Le Boiteux, V. Lorent, J. Reinhardt, and J. Baudon, 'Atomic Quantum Phase Studies with a Longitudinal Stern-Gerlach Interferometer,' Laboratoire de Physique des Lasers, Institute Galilee, Universite Paris-Nord, submitted to J. Phys. Paris II (1992).
5. We are interested in the electromagnetic coupling and its dependence on the center of mass as well as the relative coordinate. For a treatment of relativistic effects and of spin, see S. J. Brodsky and J. R. Primack, Phys. Rev. **174**, 2071 (1968).
6. The factor $c(1-c)$ is the well known 'kilroy' pattern if plotted against the angle θ, with a minimum of (-2) at $\theta = \pi$ and two symmetric maxima of $(1/4)$ at $\theta = \pi/3$ and $5\pi/3$.