Nonobservability of Early-Time Departures from Fermi's "Golden Rule"

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The problem of the early-time decay rate of a model system is addressed by examining the evolution of the probability current at a detector located a macroscopic distance from the source. If one interprets this current as the flux of decay products, early-time departures from Fermi's "golden rule" are not, in principle, observable.

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When an effectively constant interaction causes an isolated discrete level to decay into a single continuum, the amplitude of the initial state, $a_1(t)$, behaves, to a very good approximation, such that its absolute value is given by the well-known law, $|a_1(t)|=\exp(-\gamma t/2)$, where γ is the transition rate that one would calculate from Fermi's "golden rule" in first-order perturbation theory. It is also well known that the exponential law fails at early times, a feature associated with the presence of large transient amplitudes for continuum states far removed from resonance. Only if the continuum is unbounded and the density of final states and the matrix elements of the coupling are energy independent is the Wigner-Weisskopf (WW) approximation, which is used to derive the exponential decay, exact.[1]

There has recently been some work[2,3] devoted to calculating the early temporal evolution of $|a_1(t)|$, in which the nonlinearity of the time derivative is explicitly displayed. The present paper is written to explore whether this behavior really represents a departure from a constant decay rate as determined by a signal registered by a remote detector. In part, I seek the answer to the question, "What is the connection between $1-|a_1(t)|^2$ and *measurements* of the probability that the system has decayed?" I am not challenging the details of the calculations reported in Refs. 2 and 3, but questioning whether they apply to experiments that are, or could be, performed. Instead of determining $|a_1(t)|^2$, the point of view shall be adopted that a measured rate of decay is to be identified with the probability current arriving at the detector. For $t\rightarrow\infty$, this current is identical to $d[1-|a_1(t)|^2]/dt$, but the two results are in disagreement at early times. If the interpretation of the asymptotic current as "the" decay rate is correct, the conclusion will be that the "golden rule" applies even as $t\rightarrow 0$.

Consider a hypothetical experiment. We assume that at $t=0$, a system of microscopic extent, the source, is prepared in an eigenstate of the unperturbed Hamiltonian, H_0, and the perturbation, H', is switched on. This could correspond either to a situation where the interaction is always present, and the system suddenly projected into an eigenstate of H_0, or to a case where the system entered the initial state at an earlier time, with the interaction, represented, e.g., by a classical field, suddenly turned on. A transition to the continuum (the decay) is recorded via a detector placed at a macroscopic (i.e., asymptotic) distance from the source. If the decay product is a particle of velocity v reaching the detector at time t_d, it is implicit that the decay occurred at an earlier time $t_e=t_d-r/v$. The experimental decay law is determined by performing an ensemble average of $n(t_e)$, where n indicates the relative number of particles emitted at times t_e.

The foregoing suggests that the theoretical description of the decay rate that best corresponds to experiment is, in fact, given by the surface integral of the probability current density over a sphere of radius r. We shall explicitly calculate, in the position representation, the time evolution of the wave function $\psi(t)$ for a model problem, noting that only its behavior at large r is of significance.

Whether or not the law of constant initial decay rates is rigorously true seems to be independent of the details of the system under investigation, and we should be able to deduce global properties of discrete-continuum transitions within the context of a specific calculation, so long as the problem selected for solution does not contain unrepresentative artificialities. With this restriction, one is free to analyze any convenient model system. We choose for the "decay" process the photodetachment induced in a model negative ion by a classical radiation field. The light is turned on abruptly at $t=0$, and maintained at constant amplitude thereafter. We shall work in the context of the dipole and rotating-wave approximations. The first is adopted merely for convenience—it will be obvious that the conclusions drawn will apply to an arbitrary combination of multipoles. We shall see later that making the rotating-wave approximation has no observable consequences in the present picture.

In this model, a particle initially bound in an s state by a local potential interacts with an external quasiharmonic electric field polarized along the z axis. The field is given by $E(t)=2E_a \cos\Omega t$, with $E_0(t)=2E_a S(t)$, with $S=0$, $t<0$, and $S=1$, $t>0$. Our task is to solve the time-dependent Schrödinger equation, and determine the asymptotic form of the wave function and the probability current density. It will be sufficient to work in first-order perturbation theory, since the lowest nonvanishing contribution to the current will be proportional to E_a^2.

The full wave function may be expanded as a power series in E_a, yielding $\psi(t)=\sum_{n=0}^{\infty}E_a^n\psi_n(t)$. The $n=1$ contribution is the solution to

$$(H_0-i\,\partial/\partial t)\psi_1=-H'\psi_0(\vec{r},t)=-S(t)r\cos\theta[u_0(r)/r]\exp[-i(\omega_0+\Omega)t]Y_0^0(\theta,\phi), \tag{1}$$

where $H_0=-\nabla^2/2+V(r)$, $[u_0(r)/r]Y_0^0(\theta,\phi)$ is the spatial part of the unperturbed wave function, and ω_0 is the initial-state unperturbed energy. We work in atomic units here and throughout the paper. It is not necessary to calculate $\psi_2(t)$, since the contribution to the current arising from the cross term between ψ_2 and ψ_0 is exponentially small for $r\rightarrow\infty$. The right-hand side of Eq. (1) transforms like $P_1(\cos\theta)$, so that the left-hand side must also be of the form $u_1(r,t)\cos\theta/r$, with u_1 satisfying

$$\left[-\frac{1}{2}\frac{d^2}{dr^2}+\frac{1}{r^2}+V(r)-i\frac{\partial}{\partial t}\right]u_1(r,t)=-\frac{Sru_0(r)}{(4\pi)^{1/2}}\exp[-i(\omega_0+\Omega-i\epsilon)t]. \tag{2}$$

The factor $\exp(-\epsilon t)$ is appended, as usual, to make a transformation to the frequency domain convergent. It is understood in the following that passage to the limit $\epsilon\rightarrow 0$ is always implied.

Writing $u_1(r,t)$ in terms of its Fourier transform, we have

$$u_1(r,t)=(2\pi)^{-1/2}\int_{-\infty}^{\infty}\tilde{u}_1(r,\omega)\exp[-i(\omega+i\epsilon)t]\,d\omega,$$

where $\tilde{u}_1(r,\omega)$ satisfies

$$\left[-\frac{1}{2}\frac{d^2}{dr^2}+\frac{1}{r^2}+V(r)-\omega-i\epsilon\right]\tilde{u}_1(r,\omega)=\frac{-i}{\sqrt{2}2\pi}\frac{ru_0(r)}{\omega-\omega_0-\Omega+i\epsilon}. \tag{3}$$

The inhomogeneity is the Fourier transform of the right-hand side of Eq. (2). Equation (3) may be formally integrated via the Green's function $G_\omega(r,r')$, which is a solution of

$$\left[\frac{d^2}{dr^2}-\frac{2}{r^2}-2V(r)+k^2+i\epsilon\right]G_\omega(r,r')=\delta(r-r'), \tag{4}$$

where $k^2=2\omega$. In the region of interest, $r\rightarrow\infty$ and

$$G_\omega(r,r')\sim\exp(i\delta_\omega)v_1(kr')\exp(ikr),$$

where δ_ω is the $L=1$ phase shift at frequency ω, and v_1 is a solution, regular at $r=0$, of the homogeneous version of Eq. (4) that behaves asymptotically like $\sin(kr-\pi/2+\delta_\omega)/k$. Accordingly, the asymptotic form of $\tilde{u}_1(r,\omega)$ is

$$\tilde{u}_1(r,\omega)\sim+\frac{i}{\sqrt{2}\pi}\frac{\exp(i\delta_\omega)M_\omega\exp(ikr)}{\omega-\omega_0-\Omega+i\epsilon},$$

where $M_\omega=\int_0^\infty r'u_0(r')v_1(kr')\,dr'$. Returning to the time domain, we have

$$u_1(r,t)=+\frac{i}{2\pi^{3/2}}\int_{-\infty}^{\infty}\frac{\exp[-i\omega t-kr+i\epsilon t)]\exp(i\delta_\omega)M_\omega\,d\omega}{\omega-\omega_0-\Omega+i\epsilon}. \tag{5}$$

The right-hand side of Eq. (5) bears a close resemblance to the integral appearing in the description of the dynamics of a free wave packet,[1] and we express the exponent in terms of the resonant frequency $\omega_r=\omega_0+\Omega$, with $k_r^2=2\omega_r$. This yields

$$u_1(r,t)=\frac{+i}{2\pi^{3/2}}\exp[-i(\omega_r t-k_r r+i\epsilon t)]I,$$

where

$$I=\int_{-\infty}^{\infty}\frac{\exp\{-i[(\omega-\omega_r)(t-r/k_r)+2(\omega-\omega_r)^2r/k_r(k+k_r)^2]\}}{\omega-\omega_r+i\epsilon}M_\omega\exp(i\delta_\omega)\,d\omega.$$

The contributions to I arising from the quasi Gaussian and the denominator are very sharply peaked around ω_r. Accordingly, we replace the factor $M_\omega\exp(i\delta_\omega)$ by its resonant value and remove it from the integrand, yielding

$$I=M_{\omega_r}\exp(i\delta_{\omega_r})\int_{-\infty}^{\infty}\frac{\exp[-i(\omega-\omega_r)(t-r/k_r)]\exp[-2i(\omega-\omega_r)^2r/k_r(k+k_r)^2]\,d\omega}{\omega-\omega_r+i\epsilon}=M_{\omega_r}\exp(i\delta_{\omega_r})J.$$

This *Ansatz,* is, of course, identical to that made in deriving the "golden rule." The difference is that in the present case we are able to estimate the error introduced into the probability current by the approximation. We shall return to this point later, and comment now merely that the correction to ψ_1 associated with the replacement is $O(r^{-1/2})$, which does not contribute to the current in the asymptotic region. The remaining integral is of the form of the Fourier transform of the product of two functions, which may be expressed in the time domain by the *Faltung* theorem. This is

$$J=-(2\pi)^{1/2}i\int_{-\infty}^{t-r/k_r}F_G(t')\,dt',$$

where the effect of the transform of the denominator, a temporal step function, is incorporated into the limits, and F_G is the Fourier transform of the quasi Gaussian. The function J is very nearly a step function—it is vanishingly small until $t\sim r/k_r-s$, where s is a time $\sim(r/k_r^3)^{1/2}$, short compared to r/k_r, and is essentially unity after $t\sim r/k_r+s$. The exact form of J is not amenable to computation, but its features may be understood by approximating the quasi Gaussian by a true Gaussian. In this approximation, which is valid with an error that is asymptotically small, since $r>>1$,

$$J\simeq-i(\pi k_r^3/r)^{1/2}(1-i)\int_{-\infty}^{t-k_r/r}\exp(it'^2k_r^3/2r)\,dt'.$$

If the "resolving time" of the detector is $>>s$, J will be a good approximation to a step function. The temporal evolution of the probability current on the detector sphere is now clear. The current is zero for times less than $\sim r/k_r$, and rises, in an interval on the order of s, to a value given by the "golden rule." Apart from the temporal distortion of the leading edge and the transit time delay, the current follows the time evolution of the driving pulse. Note that the nonrectangular shape of the leading edge is not a consequence of the breakdown of the WW approximation, but occurs as a result of the asymptotic energy-momentum dispersion relation. The distortion is analogous to the spreading of a wave packet and would be present even if WW were exact.

We return now to the approximation of replacing the matrix-element factor by its value at ω_r. The error term is

$$I_E=\int_{-\infty}^{\infty}[M_\omega\exp(i\delta_\omega)-M_{\omega_r}\exp(i\delta_{\omega_r})]\exp\left(-\frac{2i(\omega-\omega_r)^2r}{k_r(k+k_r)^2}\right)\exp\left[-i(\omega-\omega_r)\left(t-\frac{r}{k_r}\right)\right](\omega-\omega_r+i\epsilon)^{-1}d\omega.$$

By construction, the square bracketed factor vanishes near $\omega=\omega_r$, and removes the near singularity due to the denominator. Because r is very large, the integrand oscillates wildly, and may be evaluated approximately by stationary phase. The extremum of the exponent occurs at roughly $\omega-\omega_r=-(t-r/k_r)k_r^3/r$ and gives $I_E\sim qr^{-1/2}$, where $q=k_r^{3/2}$, which is asymptotically negligible. Thus, if it is correct to identify the probability current with the experimental decay rate, early time departures from the "golden rule," arising from corrections to the WW approximation, are not observable.

To illustrate the order of magnitude of representative numerical values, consider a typical photodetachment experiment. Choose for convenience a final-state energy to be 1 Ry, so that $k_r\sim1$ a.u. Typical detachment cross sections are $\sim10^{-17}$ cm^2, so that if the photon flux is 10^5/sec (an optical intensity of a few milliwatts per square centimeter), we find a decay time $\tau\sim10^2$ sec. If the detector is assumed to be located ~1 m from the source, the transit time t_T is on the order of 1 μsec, while t_r, the rise time of the detector current associated with the distortion of the leading edge, is $\sim10^{-12}$ sec.

Thus, the signal is delayed with respect to the onset of the interaction by a time virtually identical to t_T, while t_r is $\sim 10^{-6}$ of t_T. Both t_T and t_r are very short compared to and independent of τ.

I note that if one applied the same procedure to calculate the contribution to the wave function due to the antirotating component of the radiation field, one would find an exponentially decaying function of r in the asymptotic region, which likewise makes a negligible contribution to the current. Similarly, the "reversible ionization" reported by Haan and Geltman[4] would not be observable if the decay rate is to be identified with the current. Those authors analyzed three-photon ionization, and found contributions to $1-|a_1(t)|^2$ due to below-threshold (one- and two-photon absorptions) continuum-state amplitudes. The components of the wave function associated with those amplitudes are also asymptotically exponentially decreasing functions of r, and make no measurable contribution to the current.

If the interaction is switched on in a short, but finite, time, the results will be essentially unaffected, since effects will be masked by the distortion of the leading edge of the pulse. This stands in distinction to the large-time behavior, which is significantly modified, a result presented recently by Mittleman and Tip.[5]

The present analysis is based on the notion that the decay is detected at macroscopic distances, and may not be applicable in experiments where the detector is very close to the decaying system. Furthermore, it assumes that the system undergoing transitions is accurately an isolated discrete state coupled to a true continuum. Clearly, if the final state is quasidiscrete the transition rate may exhibit nonexponential behavior at all times.

To summarize, it is found that if one associates the decay rate of a system with the probability current in the asymptotic region, early-time deviations from the "golden rule" arising from the breakdown of the Wigner-Weisskopf approximation are not detected. Nonlinearities in the time derivative of $|a_1|$ manifest themselves in the probability flux only in the near field—they are absent in the asymptotic region and do not contribute to the recorded decay. With this point of view, the "golden rule" is valid from the earliest onset of the signal, except for an interval during which the current grows to its steady value. This finite–rise-time effect, which is related to wave-packet spreading, would be present even in cases where the Wigner-Weisskopf approximation holds exactly.

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[1]E. Merzbacher, *Quantum Mechanics* (Wiley, New York, 1970).

[2]A. M. Lane, Phys. Lett. **99A**, 359 (1983).

[3]S. Geltman, J. Phys. B **10**, 831 (1977).

[4]S. L. Haan and S. Geltman, J. Phys. B **15**, 1229 (1982).

[5]M. H. Mittleman and A. Tip, J. Phys. B **17**, 571 (1984).