Generalisation of Fermi's golden rule for the strong bound-free transitions

1989 J. Phys. A: Math. Gen. 22 4163

(http://iopscience.iop.org/0305-4470/22/19/011)

J. Phys. A: Math. Gen. 22 (1989) 4163-4169. Printed in the UK

Generalisation of Fermi's golden rule for the strong bound-free transitions

Rafał J Kukliński and Kazimierz Rzążewski

Institute for Theoretical Physics, Polish Academy of Sciences, Al Lotników 32/46, 02-668 Warsaw, Poland

Received 27 April 1988

Abstract. A simple analysis of the bound-free system shows that for a narrow continuum the Fermi golden rule can be generalised for the high-intensity regime. This generalisation has the form of a simple expression for the decay rate and helps to explain nearly perfect population trapping in the case of a narrowly peaked matrix element. Such population trapping means that the decay rate of a bound state strongly coupled to a continuum tends to zero for increasing coupling strength only if the matrix element falls off faster than the Lorentzian. We also find new solutions for pulsed excitation giving exact population trapping in the bound-free transition.

Quantum mechanical systems often undergo transitions from a bound state to a continuum. Such transitions, caused either by static or time-dependent perturbation, are responsible for decay processes in quantum physics. On the other hand, when the continuum has a very 'narrow' structure, i.e. the energies of continuum states vary only within a narrow band, the bound-free system becomes similar to a two-level system. The example of autoionisation resonance shows that for strong enough coupling to a 'narrow' continuum the exponential decay of the bound state may be accompanied by 'beats' of population, similar to Rabi oscillations (Fano 1961). Considering the band structures in solids, we deal with almost 'rectangular' matrix elements describing the interaction with the continuum (ionisation thresholds in negative ions give only a one-side cut-off of the matrix element). Also interactions with optical phonons give examples of narrow continuum structure, where the matrix element falls to zero far from the resonant centre (see, for example, von Foerster and Glauber 1971). It is obvious that in the limit of small width of the continuum we deal with a two-level atom. The same similarity can be found when we increase the coupling strength for a given and constant width of the continuum ('saturation of the continuum'). As we mentioned, such an effect was found for autoionisation, described with the help of a bound-free system with a Lorentzian-shaped matrix element. The saturation phenomena for such types of coupling were discussed in detail by Cohen-Tannoudji and Avan (1977). A recent study of this system with a more rapidly decreasing Gaussian matrix element has shown a decreasing decay rate for increasing coupling strength (Rzążewski and Mostowski 1987).

In the first part of our paper we analyse the bound-free system for a set of model coupling functions giving exact non-perturbative solutions. In the second part we find a general formula for the decay rate for an arbitrarily 'narrow' matrix element in the strong-coupling limit. This formula expresses the value of the decay rate by the matrix element shifted from the energy conservation condition. Such a formulation is therefore

0305-4470/89/194163+07$02.50

similar to Fermi's golden rule, which describes the opposite, weak-coupling limit. Finally we present a solution for a time-dependent coupling in order to get a better understanding of the bound-free interaction in the strong-coupling limit. The bound-free system in the non-perturbative regime has been analysed many times via various approaches in the literature (Fano 1961, Heller and Popov 1976, Cohen-Tanoudji and Avan 1977, Lambropoulos and Zoller 1980, Andryushin *et al* 1982, Coleman and Knight 1982; see also Cohen-Tannoudji *et al* 1977). However our analysis is the first to describe the asymptotic limit for arbitrary coupling to 'narrow' continua.

We consider a quantum mechanical system consisting of one discrete level $|0\rangle$ and a continuum of states $|\omega\rangle$ with standard normalisation

$$\langle 0|0\rangle = 1 \qquad \langle\omega|\omega'\rangle = \delta(\omega-\omega').$$

The Hamiltonian of such a system consists of a free part:

$$H_0 = \hbar\omega_0|0\rangle\langle 0| + \int \mathrm{d}\omega\ \hbar\omega|\omega\rangle\langle\omega| \tag{1}$$

and an interaction:

$$H_\mathrm{I} = \int \mathrm{d}\omega\ \Omega(\omega)(|0\rangle\langle\omega| + |\omega\rangle\langle 0|) \tag{2}$$

which is time independent for the transitions caused by static perturbation. For the harmonic, time-dependent perturbations, such as in the important case of the interaction with coherent monochromatic light, the rotating-wave approximation brings the interaction to the time-independent form (2). All the energy integrals will be extended from $-\infty$ to $+\infty$, so our model can be applied sufficiently far above the threshold.

The dynamics is fully determined by the matrix element $\Omega(\omega)$. In the well studied regime of weak coupling the probability $P(t)$ of remaining in the bound state decays exponentially:

$$P(t) = \mathrm{e}^{-\Gamma t} \tag{3}$$

with the decay rate given by Fermi's golden rule (FGR; see Fermi 1950):

$$\Gamma = 2\pi|\Omega(\omega_0)|^2. \tag{4}$$

Note that the rate is determined by the local (corresponding to the energy conservation) value of the coupling. The textbook perturbative formulation of FGR deals with short times ($\Gamma t \ll 1$). For such times the decay probability increases linearly with time: $1-P(t)=\Gamma t$.

As the coupling strength increases the global properties of $\Omega(\omega)$ become relevant. Only the 'flat continuum' ($\Omega(\omega)=\lambda=$ constant) gives an exact exponential decay, with the decay rate proportional to the coupling intensity (e.g. $P(t)=\exp(-2\pi\lambda^2 t)$). For such coupling the 'photoelectron spectrum' i.e. the population of the continuum $S(\omega)=|\langle\Psi(t)|\omega\rangle|^2$ for $t\to\infty$ has an exact Lorentzian shape $S(\omega)=\lambda^2(\omega^2+(\pi\lambda^2)^2)^{-1}$.

For other forms of the matrix element, saturation phenomena occur. In such cases the probability $P(t)$ undergoes damped oscillatory motions. It has been noted only recently (Rzążewski and Mostowski 1987) that the asymptotic value of the damping depends on the way the function $\Omega(\omega)$ falls off from its centre (we assume a resonant maximum at ω_0). For fast decreasing continua the decay rate tends to zero for strong coupling, which means an almost exact 'population trapping'.

The main aim of this paper is to obtain a simple estimate of the residual decay rate of resonant, saturated, bound-free transitions.

The Schrödinger equation for our system leads to a set of integrodifferential equations for the probability amplitudes $\beta(\omega, t) = \langle\omega|\Psi(t)\rangle$ and $\alpha(t) = \langle 0|\Psi(t)\rangle$:

$$\partial_t\alpha(t) = -\mathrm{i}\omega_0\alpha(t) - \mathrm{i}\int \mathrm{d}\omega\,\Omega(\omega)\beta(\omega, t)$$
$$\partial_t\beta(\omega, t) = -\mathrm{i}\omega\beta(\omega, t) - \mathrm{i}\Omega(\omega)\alpha(t) \tag{5}$$

with the initial conditions $\alpha(0) = 1$ and $\beta(\omega, 0) = 0$.

This set of equations is easily solvable by Laplace transform if the matrix element has the form $\Omega_n(\omega) = \Omega_0 g_n(\omega)$, where g_n is (for $\int \mathrm{d}\omega |g_n(\omega)|^2 = 1$):

$$g_n = N_n \frac{1}{[(\omega - \omega_0) + (\mathrm{i}\gamma/\sqrt{n})]^n}. \tag{6}$$

The normalisation factor N_n is given by

$$N_n^2 = \frac{((n-1)!)^2(2\gamma)^{2n-1}}{2\pi(2n-2)!\,n^{n-1/2}}.$$

Note that in the limit $n \to \infty$ we obtain a Gaussian coupling.

The Laplace transform of $\alpha(t)$ takes the form

$$\tilde{\alpha}(z) = \frac{1}{\boldsymbol{F}(z)} \qquad \boldsymbol{F}(z) = z + \int_{-\infty}^{+\infty} \mathrm{d}\omega \frac{|\Omega(\omega)|^2}{z + \mathrm{i}\omega}. \tag{7}$$

To find the explicit form of $\alpha(t)$ we have to perform the integral in (7) for $\mathrm{Re}\, z > 0$ (evolution for $t > 0$) to find $\boldsymbol{F}(z)$. In the case when the matrix element has the form $\Omega(\omega) = \Omega_n(\omega)$ ('generalised Lorentzian' coupling) $\boldsymbol{F}(z)$ is just a polynomial, and therefore zeros of $\boldsymbol{F}(z)$ (e.g. pole contribution to the inverse Laplace transform; see also Weisskopf and Wigner (1930)) can be easily found, at least numerically.

The most common shape of the resonance is that of the Lorentzian type $\Omega_1(\omega)$ (we take $\omega_0 = 0$ for simplicity). In the weak-field limit ($\Omega_0 < \gamma$), we have FRG-type decay (the spectrum is Lorentzian in this case). For strong coupling ($\Omega_0 \gg \gamma$) the asymptotic behaviour for the probability amplitude $\alpha(t)$ is given by (Cohen-Tanoudji 1977, Rzążewski and Eberly 1981, Rzążewski and Mostowski 1987):

$$\alpha(t) \simeq \exp(-\Gamma t/2)\cos(\Omega_0 t). \tag{8}$$

It is seen that we have 'beats' of the probability amplitude. The saturated ($\Omega_0 \gg \gamma$) decay rate Γ is independent of the coupling strength Ω_0 and is equal to the width of the continuum γ (the frequency of beats Ω_b tends to Ω_0). In the limit $\gamma \to 0$ we recover the well known undamped Rabi oscillations.

Analysing the explicit form of $\alpha(t)$ calculated from (7) for the coupling $\Omega(\omega) = \Omega_n(\omega)$, we found that such Rabi-like oscillations appear for all n strong enough coupling e.g. the probability amplitude has the form

$$\alpha(t) \simeq \exp(-\Gamma(\Omega_0)t/2)\cos(\Omega_b(\Omega_0)t). \tag{9}$$

The coupling strength enters through the parameter Ω_0. We found that in the strong-coupling limit (e.g. $\Omega_0 \gg \gamma$) the frequency of beats Ω_b tends to Ω_0. A natural consequence of such population beats is that the 'final' population of the continuum $S(\omega) = |\langle\Psi(t \to \infty)|\omega\rangle|^2$ consist of two peaks around $\omega \simeq \pm\Omega_b(\Omega_0)$ (figure 1; see also Autler and Townes (1955)).

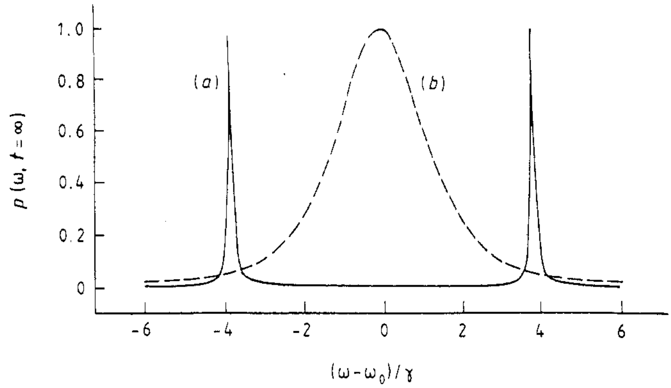

Figure 1. Population of the continuum $S(\omega)=|\langle\Psi(t\to\infty)|\omega\rangle|$ (a) plotted with the matrix element $|\Omega_2(\omega)|^2$ (b). The intensity is $\Omega_0=4\gamma$.

Analysing the decay rate $\Gamma(\Omega_0)$ we found an asymptotic behaviour $\Gamma(\Omega_0)=(\Omega_0)^{-2(n-1)}$, which means that if only $n>1$ the decay rate tends to zero (see also Rzążewski and Mostowski 1987) in the strong-coupling limit. Remarkably the decay rate must be well estimated by the square of the matrix element taken in place corresponding to the peaks in the continuum, e.g.

$$\Gamma(\Omega_0)=\chi(\Omega_0)|\Omega(\omega=\Omega_b\simeq\Omega_0)|^2. \tag{10}$$

Such a formulation may be viewed as a straightforward generalisation of the perturbative FRG. The function $\chi(\Omega_0)$ for weak couplings (perturbative regime) is simply equal to 2π. In figure 2 the ratio $x(\Omega_0)=\Gamma(\Omega_0)/2\pi|\Omega(\Omega_0)|^2$ is plotted for varying Ω_0 and different n (we take $\Omega(\omega)=\Omega_n(\omega)$).

As we can see, for strong couplings ($\Omega_0\gg\gamma$) the factor $\chi(\Omega_0)$ approaches π ($x(\Omega_0)=\frac{1}{2}$, which is a result independent of n. For square integrable couplings ($\int d\omega|\Omega(\omega)|^2<\infty$) this is a general feature which can be shown analytically.

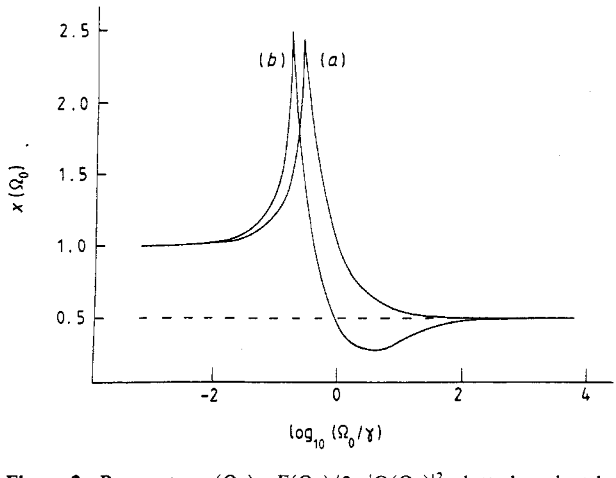

Figure 2. Parameter $x(\Omega_0)=\Gamma(\Omega_0)/2\pi|\Omega(\Omega_0)|^2$ plotted against $\log_{10}(\Omega_0/\gamma)$ for (a) $n=1$ and (b) $n=5$.

We assume that the matrix element has the form $\Omega(\omega)=\Omega_0 g(\omega)$ where $\int d\omega|g(\omega)|^2=1$. For $\Omega_0\to\infty$ the estimate of the zeros of $F(z)$ with the smallest negative real part can be done as follows. Our task is equivalent to finding zeros of $zF(z)/\Omega_0^2=(z/\Omega_0)^2+1+G(z)$ where for pure imaginary z $G(z)$ is given by

$$G(z)=\lim_{\varepsilon\to 0^+}\int d\omega\frac{z|g(\omega)|^2}{z+\varepsilon+i\omega}-1. \tag{11}$$

We found that the function $G(z)$ has the property that $|z|\ll\gamma\Rightarrow|G(z)|\ll 1$. This property allows us to solve the equation $zF(z)/\Omega_0^2=0$ perturbatively with respect to $G(z)$, only if the solution z_s ($z_sF(z_s)=0$) fulfils $|z_s|\ll\gamma$. As we will see later our solution fulfils this condition when $\Omega_0\gg\gamma$, so our method is self-consistent in the regime of strong coupling.

The zeroth-order solution is equal to $z_s^0=\pm i\Omega_0$ $((z_s^0/\Omega_0)^2+1=0)$. The first-order solution gives

$$(z_s^1/\Omega_0)^2+1=-G(z_3^0).$$

Calculating $G(z_s^0)$ from (3.2) and applying an approximation

$$\left(\frac{z_s^1}{\Omega_0}\right)^2=\left(\frac{z_s^0+\Delta}{\Omega_0}\right)^2\simeq\left(\frac{z_s^0}{\Omega_0}\right)^2+\frac{2\Delta z_s^0}{\Omega_0^2}$$

we get Δ using the well known property of distributions

$$\frac{1}{(x-i\varepsilon)}=P\left(\frac{1}{x}\right)+i\pi\delta(x).$$

The real part of Δ gives us the decay rate in the strong-coupling limit ($\Gamma(\Omega_0)/2=-\mathrm{Re}(\Delta)$):

$$\Gamma(\Omega_0)=\pi|\Omega(\Omega_0)|^2. \tag{12}$$

As we can see, in the strong-coupling regime we have a compact expression for the decay rate of a bound state coupled to a continuum, very similar to Fermi's golden rule which describes the weak-coupling regime. A simple consequence of this new rule for strong couplings is the occurrence of population trapping for matrix elements 'narrower' than the Lorentzian.

Let us generalise our model to the case of the time-dependent interaction $H_I=f(t)\int d\omega\,\Omega(\omega)(|0\rangle\langle\omega|+|\omega\rangle\langle 0|)$. The Schrödinger equation for the system has the form (we substitute $\alpha\to\alpha\exp(i\omega_0 t)$ and $\beta\to\beta\exp(i\omega_0 t)$)

$$\begin{aligned}\partial_t\alpha(t)&=-if(t)\int d\omega\,\Omega(\omega)\beta(\omega,t)\\ \partial_t\beta(\omega,t)&=-i(\omega-\omega_0)\beta(\omega,t)-if(t)\Omega(\omega)\alpha(t).\end{aligned} \tag{13}$$

Such a Hamiltonian can describe, for example, a quantum optical system decaying in the electromagnetic field of a laser pulse.

The equations (13) can be solved explicitly for the exponentially growing coupling (Kukliński and Lewenstein 1987): $f(t)=e^{t/\tau}$ (assuming that for $t=-\infty$ only the bound state is populated and $\Omega(\omega)=\Omega_n(\omega)$). Such a coupling, valid till $t=0$ describes a smooth turning on of the laser light. We perform a change of variable $t\to z=e^{t/\tau}$ (this induces $\partial_t\to z/\tau\partial_z$). Now, we expand functions $\alpha(z(t))$ and $\beta(\omega,z(t))$ into a Taylor series, e.g. $\alpha(z)=\Sigma a_n z^n$ and $\beta(\omega, z)=\Sigma b_n(\omega) z^n$. Therefore the set of equations (13) lead to the following recurrence relations for coefficients a_n and $b_n(\omega)$:

$$\begin{aligned} n/\tau a_n &= -\mathrm{i} f(t) \int \mathrm{d}\omega\, \Omega(\omega) b_{n-1}(\omega) \\ (n/\tau+\mathrm{i}(\omega-\omega_0)) b_n(\omega) &= -\mathrm{i} f(t) \Omega(\omega) a_{n-1}. \end{aligned} \tag{14}$$

We easily solve this recurrence, obtaining under the assumption $\alpha(z=0)=1$ and $\beta(\omega, z=0)=0$ the solutions

$$\begin{aligned} \alpha(z) &= \sum_{n=0}^{\infty} \frac{a_n}{n!}(-z^2)^n \\ \beta(z) &= \sum_{n=0}^{\infty} \frac{a_n}{n!} \frac{-\mathrm{i}\Omega(\omega)(-1)^n}{(2n+1)/\tau+\mathrm{i}(\omega-\omega_0)} z^{2n+1} \end{aligned} \tag{15}$$

where $a_n = \Pi(\int \mathrm{d}\omega |\Omega(\omega)|^2((2k-1)/\tau+\mathrm{i}(\omega-\omega_0))^{-1}$. Taking $\Omega(\omega)=\Omega_n(\omega)$ we obtain for $\alpha(z)$ the Taylor series of a generalised hypergeometric function ${}_{n-1}F_n(\ldots; -cz^2)$.

For $\Omega(\omega)=\Omega_2(\omega)$ our solution has the form

$$\alpha(\theta) = {}_1F_2\left\{\frac{G+1/2}{(G+1)/2, (G+1)/2}; -(\theta/2)^2\right\} \tag{16}$$

where $\theta=\tau\Omega_{0^2}$ (dimensionless 'pulse area') and $G=\gamma\tau$.

In figure 3 we show the ground-state population $p(\theta)$ obtained from (16). As we see the decay channel is fully shut down.

We can explain it analysing an 'adiabatic generalisation' of the formula (9);

$$\alpha(t) = \exp\left(-\int_{-\infty}^{t} \mathrm{d}t'\, \pi|\Omega(\Omega_0(t'))|^2\right) \cos\left(\int_{-\infty}^{t} \mathrm{d}t'\, \Omega_0(t')\right). \tag{17}$$

Note that the integral $\int \mathrm{d}t\, \pi|\Omega(\Omega_0(t))|^2$ is finite, which means an incomplete damping.

The analytic solution presented here for the time-dependent interaction shows that if the bound-free coupling increases sufficiently fast in time, the population trapping is full.

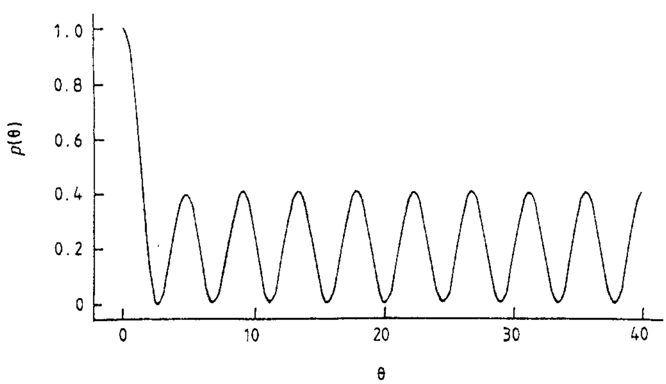

Figure 3. Population of the bound level $p(\theta)=|\alpha(\theta)|^2$ plotted against the pulse area θ for the exponential pulse.

To summarise: we have presented an analysis of the interaction of a discrete level with a narrow continuum in the highly non-perturbative regime. We have investigated the situation when the coupling strength increases for a given width of the continuum—in such a case the bound-free system becomes similar to a two-level atom because of the oscillatory behaviour of the bound-state population. Our analysis shows the existence of a well defined strong-coupling limit for the dynamics of the bound-free system, described by a general formula connecting the decay rate with the value of the matrix element shifted from the energy conservation condition. The value of this shift depends on the global structure of the matrix element and reflects the non-perturbative character of the asymptotic solution.

References

Andryushin A I, Kazakov A E and Fedorov M V 1982 *Zh. Eksp. Teor. Fiz.* **82** 91
Autler S H and Townes C H 1955 *Phys. Rev.* **100** 703
Cohen Tannoudji C and Avan P 1977 *Colloques Internationaux du CNRS* (Paris: Editions du CNRS)
Cohen Tannoudji C, Diu B and Laloë F 1977 *Quantum Mechanics* (New York: Wiley) pp 1343-50
Coleman P E and Knight P L 1982 *J. Phys. B: At. Mol. Phys.* **15** L235
Heller Yu I and Popov A K 1976 *Opt. Commun.* **18** 449
Fano U 1961 *Phys. Rev.* **124** 1866
Fermi E 1950 *Nuclear Physics* (Chicago, IL: University of Chicago Press) p 142
Kukliński J R and Lewenstein M 1987 *J. Phys. B. At. Mol. Phys.* **20** 1387
Lambropoulos P and Zoller P 1980 *Phys. Rev.* A **24** 379
Rzążewski K and Eberly J H 1981 *Phys. Rev. Lett.* **47** 408
Rzążewski K and Mostowski J 1987 *Phys. Rev.* A **35** 4414
von Foerster T and Glauber R J 1971 *Phys. Rev.* A **3** 1484
Weisskopf V and Wigner E P 1930 *Z. Phys.* **63** 54