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OSCILLATING TRANSITION PROBABILITY FOR PHOTOELECTRON EMISSION

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ABSTRACT

The time-dependent transition probability for linear photoelectron emission is calculated and the exact solution of the equation of motion is given. It shows an oscillating character of double light frequency with exponentially decreasing amplitude.

РЕЗЮМЕ

Была определена вероятность перехода линейной фотоэлектронной эмиссии с помощью точного решения уравнения движения оператора плотности. Оказалось, что вероятность перехода носит колебательный характер с двойной световой частотой и экспоненциально убывающей амплитудой.

KIVONAT

Lineáris fotoelektronmisszió átmeneti valószinüségét határoztuk meg a sürüségoperátor mozgásegyenletének egzakt megoldásával. Az átmeneti valószinüség kétszeres fényfrekvenciáju oszcillációt mutat, amelynek amplitudója időben exponenciálisan csökken.

The photon concept was introduced into physics by Einstein's interpretation of the photoelectric effect. Recent developments in the theory of this effect, however, have made it clear that its theoretical explanation does not require quantization of the electromagnetic field. All the experimental phenomena can be described by a semiclassical theory in which the electromagnetic field is treated classically, while only the matter interacting with electromagnetic radiation is treated quantum mechanically. This semiclassical theory accounts for the relationship between photocurrent and light intensity, both for linear and higher order photoelectric effects, as well as for the Einstein relation between electron energy and light frequency [1] , [2] .

The widely used photodetector model consists of a large number of completely independent atoms. Each atom has one electron, and for the sake of simplicity we consider the problem to be one dimensional. The atom has a ground state $|g\rangle$ and a set of quasi-continuous excited states $|k\rangle$, which are normalized in a length L very large compared to atomic dimensions, so that the levels approximate to a continuum when L tends toward infinity.

The atoms are illuminated by a classical, monochromatic electromagnetic field, for which the electric field is

$$E(t) = E_o \cos\omega t \qquad /1/$$

Transitions from the ground state $|g\rangle$ to any of the $|k\rangle$ states under the influence of incident light give rise to the emission of photoelectrons. The Hamiltonian for the electron interacting with the electromagnetic field is

$$H = H_o - e\,E(t)x \qquad /2/$$

where H_o is the unperturbed atomic Hamiltonian, x is the coordinate operator of the electron and e is its charge. Here only the elec-

tric dipole part of the interaction is taken into account, because atomic dimensions are very small compared to the wavelength of light. Spatial variations of light within the atom as well as retardation and magnetic effects are neglected. This approximation simplifies the calculations further.

It is convenient to go into the interaction picture, in which the interaction part of the Hamiltonian becomes:

$$V(t) = - e\, x(t)\, E(t) \qquad /3/$$

where

$$x(t) = \exp\left\{\frac{i}{\hbar} H_o t\right\} x \exp\left\{-\frac{i}{\hbar} H_o t\right\} \qquad /4/$$

The nonvanishing matrix elements of /3/ are

$$\langle k|V(t)|g\rangle = - e\, x_{k,g}\, E(t) \exp\left\{\frac{i}{\hbar} \varepsilon_k\, t\right\} \qquad /5/$$

and

$$\langle g|V(t)|k\rangle = - e\, x_{g,k}\, E(t) \exp\left\{-\frac{i}{\hbar} \varepsilon_k\, t\right\} \qquad /6/$$

where ε_k is the energy of the k -th excited state measured relative to the ground state.

For the description of the transition we use the density matrix formalism. The density matrix of photoelectrons obeys the following equation of motion in the interaction picture:

$$i\hbar\, \dot{\rho}(t) = \left[V(t),\ \rho(t)\right] \qquad /7/$$

the formal solution of which is:

$$\rho(t) = \rho(0) - \frac{i}{\hbar} \int_0^t dt' \left[V(t'),\ \rho(t')\right] \qquad /8/$$

It is convenient to substitute /8/ into /7/ to obtain a more useful form of the equation of motion for the following calculations:

$$i\hbar\, \dot{\rho}(t) = \left[V(t),\ \rho(0)\right] - \frac{i}{\hbar} \int_0^t dt' \left[V(t),\ \left[V(t'),\ \rho(t')\right]\right] \qquad /9/$$

In what follows we ignore transitions between the excited states, because we are interested in ionization process: i.e.we assume $\rho_{k,k'} \sim \delta_{k,k'}$.

Before going into the details of the calculation we briefly recall the results obtained by the time-dependent perturbation theory in lowest order on the ground of the above assumptions [1] . We begin by defining the total probability $P(t)$ of finding a photoelectron:

$$P(t) = \sum_k \rho_{k,k}(t) \qquad /10/$$

We obtain from /9/ , to the lowest order:

$$\rho_{k,k}(t) \approx \frac{2\pi}{\hbar} \frac{e^2 E_o^2}{4} |x_{k,g}|^2 \, \delta(\varepsilon_k - \hbar\omega).t \qquad /11/$$

so /10/ takes the time proportional form

$$P(t) = \gamma_1 \, t \qquad /12/$$

where

$$\gamma_1 = \frac{2\pi}{\hbar} \sigma(\hbar\omega) \frac{e^2 E_o^2}{4} |x_{k,g}|^2 \qquad /13/$$

Here $\sigma(\hbar\omega)$ is the number of states per unit energy range calculated at resonance. It should be noted that an electron will be excited on the k -th level only if $\varepsilon_k = \hbar\omega$. We denote by $A = \varepsilon_1$ the energy of the first excited state of the quasicontinuous spectrum. This is the ionization energy of the atom or the work function of the photodetector. Measuring the kinetic energy $E > 0$ of the liberated electron from the ionization energy, we obtain from the argument of the δ function in /11/ $\hbar\omega = A+E$, which is the well-known Einstein photoelectric formula. It will be noticed that /12/ is proportional to the light intensity.

In most time-dependent problems one is content with a result like /12/ . But because of the simplicity of our photodetector model it is possible to carry out the calculation to all orders in the perturbation, i.e. to integrate /9/ exactly. We begin by noting that $P(t)$, as defined in /10/ , obeys the following equation of motion taking into account /9/ :

$$\frac{dP(t)}{dt} = \sum_k \dot{\rho}_{k,k}(t) = \left(-\frac{i}{\hbar}\right)^2 \sum_k \langle k| \left[V(t), \int_o^t dt' \left[V(t'), \rho(t')\right]\right] |k\rangle \qquad /14/$$

or writing out the commutator explicitly:

$$\frac{dP(t)}{dt} = -\frac{1}{\hbar^2} \int_o^t dt' \sum_k \Big\{ V_{k,g}(t)\, V_{g,k}(t')\, \rho_{k,k}(t') + \rho_{k,k}(t')\, V_{k,g}(t')\, V_{g,k}(t) -$$

$$- V_{k,g}(t')\, \rho_{g,g}(t')\, V_{g,k}(t) - V_{k,g}(t)\, \rho_{g,g}(t')\, V_{g,k}(t') \Big\} \qquad /15/$$

Since $\rho_{g,g}(0) = 1$ /the system is initially in the ground state/ and $\rho_{g,g}(t) = 1 - \sum_k \rho_{k,k}(t)$, where all the $\rho_{k,k}(t)$ - s are positive, it is obvious that the $\rho_{k,k}(t')$ - s must have an upper bound as function of k. When L, the normalization length, tends towards infinity, the k states will approximate to a continuum. Only the integral of $\rho_{k,k}(t)$ over a finite range of energy gives rise to P(t). Thus as $L \to \infty$ the terms in /15/ involving $\rho_{k,k}(t')$ can be neglected in comparison to those involving $\rho_{g,g}(t')$, so from /15/ there remains

$$\frac{dP(t)}{dt} = \frac{1}{\hbar^2} \int_o^t dt' \sum_k |e\, x_{k,g}\, E_o|^2 \cos\omega t \cos\omega t'\, \rho_{g,g}(t')\, 2\cos\frac{\varepsilon_k}{\hbar}(t-t') \qquad /16/$$

Replacing the sum over k by an integral over excited state energies according to $\sum_k \ldots = \int d\varepsilon\, \sigma(\varepsilon) \ldots$, where $\sigma(\varepsilon)$ has the same meaning as in /13/, and using the fact that $\rho_{g,g}(t') = 1 - P(t')$, we obtain:

$$\frac{dP(t)}{dt} = \frac{1}{\hbar^2} \int_o^t dt' \int d\varepsilon\, \sigma(\varepsilon) |e\, x_{k,g}\, E_o|^2 \cos\omega t \cos\omega t' \left[1 - P(t')\right] 2\cos\frac{\varepsilon}{\hbar}(t-t') \qquad /17/$$

The integration over ε leads to the delta function $2\pi\hbar\delta(t - t')$, and after performing the t' integration we finally end up with the simple differential equation:

$$\frac{dP(t)}{dt} = \gamma\left(1 + \cos 2\omega t\right)\left(1 - P(t)\right) \quad , \qquad /18/$$

where

$$\gamma = \frac{2\pi}{\hbar} \frac{e^2 E_o^2}{2} |x_{k,g}|^2 \sigma(\hbar\omega) \qquad /19/$$

Apart from a numerical factor, /19/ is equal to /13/ .

The solution of the differential equation /18/ for the initial condition $P(0) = 0$

$$P(t) = 1 - \exp\left\{-\left(\gamma t + \frac{\gamma}{2\omega} \sin 2\omega t\right)\right\} \qquad /20/$$

This solution differs from that found by Lamb [1] in the oscillating exponential term. To understand better its physical meaning, let us expand it as a power series in $\gamma (\gamma t << 1)$, taking into account only the first order term:

$$P^{(1)}(t) = \gamma t + \frac{\gamma}{2\omega} \sin 2\omega t \qquad /21/$$

This expression can be identified with the lowest order transition probability $W^{(1)}_{k,g}(t)$, and its time derivative is the probability of transition per unit time:

$$w^{(1)}_{k,g}(t) = \frac{dW^{(1)}_{k,g}(t)}{dt} = \gamma(1 + \cos 2\omega t) \qquad /22/$$

This means physically that the transition probability, and hence the photocurrent, is an oscillating function of time. It oscillates at double light frequency around the value obtained in lowest order perturbation theory. Eq. /20/ shows how the oscillation develops in time. The amplitude of the oscillating term exponentially decreases with a time constant $\tau = \gamma^{-1}$.

We conclude that there may exist a modulation of the photocurrent, but this theoretical prédiction needs more experimental support. We hope to return to the problem in a subsequent paper that will take into account multiphoton transitions and the transitions between excited states.

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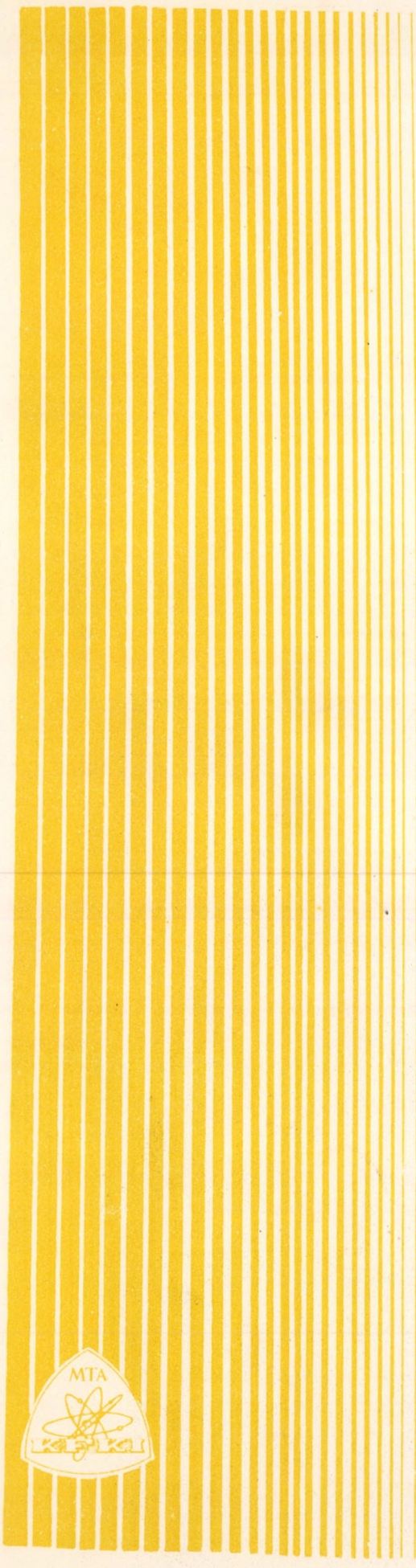

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