Pumping and acceleration of a quantum system in external fields when there are internal transitions

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We consider the pumping of a quantum system (atom, group of atoms, molecule) in external fields or coupling fields through internal transitions under the influence of a resonance field which causes internal transitions of the system from one level to another and vice versa. Such transitions change the interaction of the system with an external field (changing the polarizability, dipole moment, and so on) and causes pumping of the system as a whole. We show that by varying the strength $E_0(t)$ of the resonance field we can change the frequency of this pumping within wide limits: $\Omega(t) = 2d_{12}E_0(t)/h$; this facilitates the pumping in the case of a non-linear external coupling which requires the frequency to change with the level of excitation. We show that the excitation of internal transitions can guarantee a continuous acceleration in alternating potential fields due to the change in the system parameters at the required time intervals or at the required spatial intervals.

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INTRODUCTION

Recently, starting with [1,2], there have been intensive studies of the possibilities of a selective multi-stage pumping of atoms or groups of atoms in molecules in external fields by means of powerful coherent radiation. In view of the fact that in many cases the pumping coupling is non-linear while the broadening of the resonance frequencies by the field [1,2] does not guarantee the suppression of the anharmonicity, one must for an effective, fast and strong excitation specify and realize a fast change with time of the exciting frequency, [1] or apply a set of resonance frequencies, [3] or change the resonance frequency of the coupling through external agents, [4] which is not always convenient and effective.

We study in the present paper a method proposed in [3] for pumping a quantum system (atom, group of atoms, micro-particle) in an external field or a coupling field in a molecule by the excitation of internal transitions in a quantum system. The change of the pumping frequency of the system in an external field can then be guaranteed simply by changing the field amplitude which accomplishes the internal resonance transitions.

1. INTERNAL TRANSITIONS

We assume that the quantum system (atom, molecule, and so on) has electronic or vibrational levels and is excited by an external field $E_0 \sin \omega t$ with a frequency ω which is close to the frequency of the transition between the levels 1–2: $\omega_0 = (\mathscr{E}_2 - \mathscr{E}_1)/h$ while transitions to other levels are either impossible or have a low probability. The behavior of the system can in that case be described rather well by a two-level system in a resonance field (see, e.g., [5]). Such a system performs transitions from the one level to the other and vice versa with a frequency

$$\Omega = \{(\omega-\omega_0)^2 + (2d_{12}E_0/h)^2\}^{1/2} \approx 2d_{12}E_0/h$$

when the mismatch is small $\omega \to \omega_0$. Here d_{12} is the dipole matrix element of the transition between the levels, ω_0 is the resonance frequency of the transition. Normally, the frequency Ω for migration from one level to the other and vice versa is much less than the frequency ω_0 of the resonance transition, $\Omega \ll \omega_0$. We see that the migration frequency can easily be changed by a change in the amplitude $E_0(t)$ of the resonance field.

If the system has different interaction potentials in the external field in the different states the internal transitions can be used for an external pumping of the system.

When $\omega_0 \gg \Omega$ we can neglect the change in the frequency ω_0 when the system moves in external fields E_{ext} because of the small polarizability of the internal states. Indeed, [5] as $\Delta\omega_0^{(1)} = 0$,

$$\Delta\omega_0 \approx \Delta\omega_0^{(2)} \approx \sum{}' |V_{mn}|^2/(\mathscr{E}_m - \mathscr{E}_n) \approx |d_{12}E_{ext}|^2/\hbar^2\omega_0.$$

and the resonance broadening $\Delta\omega_{res} \approx \Omega \approx d_{12}E_0/h$ so that the drift of the frequency ω_0 due to the motion of the system in the external field is less than the width of the resonance line:

$$\Delta\omega_0/\Omega \approx (\Omega/\omega_0)(E_{ext}/E_0)^2 \ll 1,$$

which is satisfied even when $E_{ext} \sim E_0$.

We estimate the necessary resonance fields for the excitation of the internal transitions which are sufficient for the realization of the frequencies of the external oscillations: $E_0 = h\Omega/2d_{12}$. If one needs to achieve pumping in the infra-red band with $\Omega \approx 10^{13}$ to 10^{14} s^{-1}, we need, when $d \approx 10^{-18}$ in SI units, $E_0 \approx 5 \times 10^3$ to 5×10^4 in SI units, and then the resonance frequency of the internal transitions can be guaranteed to satisfy $\omega \gg \Omega$ in electron or vibrational transitions. To excite oscillations in the coupling field at the surface [3] one needs $\Omega \approx 10^{12}$ to 10^{13} s^{-1}, i.e., fields $E_0 \sim 5 \times 10^2$ to 5×10^3 in SI units, and for molecular vibrations or frequencies in external fields

$$\Omega \approx \{K/M\}^{1/2} \approx \{U_{0\,max}/L^2M\}^{1/2},$$

where the coupling energy in the field $U_0 \approx \alpha E_{ext}^2$ for a polarizable particle with a polarizability $\alpha \approx a^3$, a the size of the particle, and L the width of the potential well. In that case

$$\Omega \approx E_{ext}/L\sqrt{\rho}, \quad E_0 \approx hE_{ext}/2d_{12}L\sqrt{\rho}, \quad \rho \sim M/a^3.$$

In the case of a dipole external coupling

$$U_0 \approx PE_{ext}, \quad \Omega \approx (PE_{ext}/M)^{1/2}/L, \quad E_0 \approx h(PE_{ext}/M)^{1/2}/2d_{12}L.$$

In these cases the necessary E_0 may be small.

2. BUILDUP OF OSCILLATIONS OF THE SYSTEM AS A WHOLE IN EXTERNAL FIELDS WHEN THERE ARE INTERNAL TRANSITIONS

We assume that in the different internal states 1 and 2 the system has different polarizabilities or dipole moments, i.e., different interaction potentials in an external field, $U_1(x)$ and $U_2(x)$. One can split off the time-dependence, writing for the total potential:

$$U=U_1\cos^2\Omega t+U_2\sin^2\Omega t=\tfrac{1}{2}(U_1+U_2)+\tfrac{1}{2}(U_1-U_2)\cos 2\Omega t$$
$$=U_{\text{eff}}(x)+u(x)\cos 2\Omega t.$$

This shows that the excitation is equivalent to an excitation under the action of an additional term $u(x)\cos 2\Omega t$ (equivalent to a force $F \approx \nabla u(x)\cos 2\Omega t$) in the field $U_{\text{eff}}(x)$.

If U_1 and U_2 differ little the form of U_{eff} is the same as that of U_1, and $u \ll U_1$ and can be assumed to be a small perturbation. If both potentials are parabolic:

$$U_1=K_1(x-x_1)^2, \quad U_2=K_2(x-x_2)^2,$$

we have also

$$U_{\text{eff}}=\frac{K_1+K_2}{2}\left\{x-\frac{x_1K_1+x_2K_2}{K_1+K_2}\right\}^2+C$$

as a parabolic potential while their difference u(x) can be either parabolic or linear.

In the general case the U are not parabolic, and if the levels of the system in the field U_{eff} are given, we can study the pumping regime, giving the expansion

$$u(x)\approx\alpha+\beta x+\gamma x^2+\dots .$$

We restrict ourselves to begin with to the linear terms of the expansion. The coefficient $\beta = F_0$ is the amplitude of the force of frequency 2Ω. If this frequency approaches the resonance frequency of the external coupling $2\Omega \to \Omega_0$, we get in the case of a harmonic external coupling (quantum harmonic oscillator with a parabolic form of U_{eff}) the probability for the excitation of the n-th level:

$$W_n\approx(1/n!)\,(\mathscr{E}/h\Omega)^n\exp\{-\mathscr{E}/h\Omega_0\},$$

where $\mathscr{E}$ is the work done by the external force on the oscillator:

$$\mathscr{E}=\left(\int F_0\,dt\right)^2/M\approx(F_0t)^2/M$$

when the frequencies are the same.

In the case of an anharmonic external coupling the passage through the levels can be guaranteed by varying the frequency $2\Omega(t)$ with time (this method was proposed in [1]). In our case such a change is realized by decreasing $E_0(t)$ or by the successive inclusion of the resonance frequencies

$$2\Omega=\Omega_{01},\ \Omega_{02},\ \Omega_{03}\ \text{etc., where}\ \Omega_{0k}=(\mathscr{E}_{k+1}-\mathscr{E}_k)/h$$

and so on, $\mathscr{E}$ is the energy of the system in the external field. (Such variations in the frequencies correspond to stepwise, ladderwise decreases in the amplitude E_0 of the resonance excitation of the internal vibrations.)

The case $u \sim \gamma x^2$ corresponds to the case of the modulation of the hardness of the oscillator. In the case of a classical oscillator it reduces very simply to the equation

$$\ddot{x}+\Omega_0^2\left\{1+\frac{\gamma}{\Omega_0^2}\cos 2\Omega t\right\}x=0.$$

We consider the case which is close to strong pumping: $2\Omega = \Omega_0 + \Delta$ and substituting in the previous equation a solution of the form [6]

$$x=A(t)\cos(\Omega_0+\Delta/2)t+B(t)\sin(\Omega_0+\Delta/2)t,$$

we get for the growth of the amplitudes $A \sim B \sim e^{st}$, where

$$s^2=\tfrac{1}{4}[(\gamma/2\Omega_0)^2-\Delta^2],$$

i.e., we have $s = \gamma/4\Omega_0$ as $\Delta \to 0$.

The excitation of low-frequency oscillations by high-frequency ones can turn out to be useful when one uses lasers in the appropriate band to excite internal transitions which guarantee buildup of oscillations of the system as a whole with smaller frequencies.

When there are internal transitions it is possible to excite the vibrations of atoms in molecules, at the surface, [3] in external fields to select, separate, or simply heat the material. It is possible to use the method considered here to separate isotopes.

One can use the induced migration in atoms and molecules at the surface of a substance for pumping and the emission of powerful hypersound [3] when there are shocks at the surface with a pressure amplitude $p_s \sim N_1Mv\Omega$, where N_1 is the number of atoms per cm^2 of the surface, M the mass of an atom, v the velocity, and Ω the frequency of the oscillations on the surface.

3. CONTINUOUS ACCELERATION IN POTENTIAL FIELDS WHICH ARE MANY TIMES REPEATED IN SPACE

It is well known that a particle can not pick up energy monotonically in potential fields with an alternating polarity. However, if the particle parameters—charge, dipole moment, polarizability, and so on—could change with time, continuous acceleration can be realized.

In our case internal transitions of the system can change the polarizability of the dipole moment in the required time interval and continuous acceleration may be realized through a multi-periodic potential field. We assume that the potential of the system on the external fields $U = U_0\sin^2 kz$ is given, where in the case of the polarizability $U_0 = \alpha E_{\text{ext}}^2$. The equation of motion of the system as a whole is then

$$M\ddot{z}=\alpha(t)E_{\text{ext}}^2k\sin 2kz.$$

The solution z(t) of the equation can determine the form of $\alpha(t)$ which is necessary for continuous acceleration. For instance, we give $\alpha(t) = \alpha_0\sin 2kz$. Multiplying both sides of the equation of motion by $\dot{z}$ and integrating we get

$$\frac{1}{2}M(v^2-v_0^2)=\alpha_0E_{\text{ext}}^2k\int_0^z\sin^2 2kz\,dz=\frac{1}{4}\alpha_0E_{\text{ext}}^2k\left(z-\frac{\sin 4kz}{4k}\right),$$

i.e., when $z \gg 1/4k$

$$\tfrac{1}{2}Mv^2\approx\tfrac{1}{4}\alpha_0E_{\text{ext}}^2kz$$

and the asymptotic behavior

$$\dot{z}\approx(\alpha_0E_{\text{ext}}^2k/2M)^{1/2}z^{1/2}, \quad z(t)\approx\alpha_0E_{\text{ext}}^2kt^2/8M$$

when

$$\alpha(t)\approx\alpha_0\sin(\alpha_0E_{\text{ext}}^2k^2t^2/4M),$$

i.e., a change in the resonance field

$$E_0(t)=h\Omega/2d_{12}=\frac{h}{2d_{12}}(\alpha_0E_{\text{ext}}^2k^2t/4M).$$

In the case of the acceleration of a modulated dipole moment $P(t) = P_0\sin kz$ we get $U \approx P(t)E_{\text{ext}}(z)$ and when $E_{\text{ext}}(z) = E_{\text{ext}}\cos kz$

$$E_0 \approx \frac{h}{4M_0 d_{12}} P_0 E_{ext} k^2 t.$$

We note that the action which changes the internal state can vary not only with time, but also in space, being localized in well-defined sections of the trajectory.

The continuous acceleration is the analogue of the continuous acceleration of a plasma and of plasma bunches, considered in [7].

Accelerated atoms can find a wide range of applications in plasma physics (diagnostics, populations) for "hot" chemistry. For the external fields one can use standing light waves or microwaves, electrostatic fields with an alternating polarity, and so on.

[1] G. A. Askar'yan, Zh. Eksp. Teor. Fiz. **46**, 403 (1964) [Sov. Phys.-JETP **19**, 273 (1964)].
[2] G. A. Askar'yan, Zh. Eksp. Teor. Fiz. **48**, 666 (1965) [Sov. Phys.-JETP **21**, 439 (1965)].
[3] G. A. Askar'yan and V. A. Namiot, ZhETF Pis. Red. **21**, 646 (1975) [JETP Lett. **21**, 305 (1975)].
[4] M. M. T. Loy, Phys. Rev. Lett. **32**, 814 (1974).
[5] L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics) Fizmatgiz, 1963 [Pergamon Press, 1965].
[6] L. D. Landau and E. M. Lifshitz, Mekhanika, Nauka, 1975 [Pergamon Press].
[7] G. A. Askar'yan, Zh. Eksp. Teor. Fiz. **36**, 619 (1959) [Sov. Phys.-JETP **9**, 430 (1959)].

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