INTERACTION OF ELECTRICALLY NEUTRAL OSCILLATORS WITH AN INHOMOGENEOUS RF FIELD

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The interaction between a two-level molecule and the field of a standing transverse wave of resonance frequency is considered. It is shown that conditions exist for which the molecular energy increases at the expense of the field energy. As the result of a nonlinear treatment, the internal energy and translational energy of the molecule are determined.

As is well known[1,2], a charged particle, placed in an inhomogeneous RF field, is acted upon by a force which is quadratic in the field amplitude and leads to deflection of the particle away from regions of higher field intensity:

$$\langle \mathbf{f} \rangle = -\frac{e^2}{8M\omega^2}\frac{d}{d\mathbf{r}}|\mathbf{E}|^2$$

(e is the charge, M is the particle mass, ω is the frequency, and $|\mathbf{E}|$ is the field amplitude). This force is increased by a factor of $[1 - \Omega/\omega]^{-1}$ if the charged particle is an oscillator of natural frequency Ω close to the resonance frequency of the field. The possibility of this effect in the case when the oscillator is formed by a charged particle in a magnetic field was pointed out in[3,4], and the limiting case $\omega = \Omega$, when it is not possible to use the averaging method, was examined in[5].

In the present work it will be shown that the effect investigated in[3-5] may also take place in the case of an uncharged particle possessing a dipole moment. We shall proceed from the general formula for the force acting on a dipole in an inhomogeneous electric field[6]

$$\mathbf{F} = \nabla(\mathbf{dE}) = (\mathbf{d}\nabla)\mathbf{E} + [\mathbf{d}\,\mathrm{rot}\,\mathbf{E}], \qquad (1)^*$$

and as the dipole system we shall consider a two-level molecule. The equation describing the interaction of a two-level system with an electric field has the form[7-9]

$$\ddot{\mathbf{d}} + \Omega^2\mathbf{d} = \frac{2d_0}{\hbar}(d_0^2\Omega^2 - \Omega^2\mathbf{d}^2 - \dot{\mathbf{d}}^2)^{1/2}\mathbf{E}, \qquad (2)$$

where $d_0 = \max|\mathbf{d}|$, Ω is the resonance frequency of the molecule, and $\hbar$ is Planck's constant.

First we shall assume that the field has the form of a traveling wave: $E_x = E_0 \sin\Omega(t - z/c)$, and seek the solution of (2) in the form $d_x = d_0 a(t)\cos\Omega(t - z/c)$. Then, for the amplitude $a(t)$ of the dipole moment and the molecular velocity $\dot{z}$ (taking account of (1)) we obtain the following system of equations

$$\dot{a} = -\frac{d_0E_0}{\hbar}(1-a^2)^{1/2}, \quad \ddot{z} = -\frac{d_0E_0\Omega}{Mc}a(t). \qquad (3)$$

According to (3), the amplitude of the dipole moment changes with time according to the law $a = \sin(d_0E_0t/\hbar)$ and the difference of the populations of the levels $w = (1 - a^2)^{1/2} = -\cos(d_0E_0t/\hbar)$ periodically assumes positive values (as also in the case of a homogeneous RF field[10]). At the same time the particle is acted upon by an average (over the period of the RF field) force:

$$-\frac{d_0E_0\Omega}{c}\sin\frac{d_0E_0}{\hbar}t,$$

as a result of which the molecule can be accelerated.

However, comparing the dimensionless parameters $q = d_0E_0/\hbar\Omega$ and $\mu = d_0E_0/Mc^2$, which respectively characterize the change in the internal energy of the molecule and its acceleration, we arrive at the conclusion that the latter effect is insignificantly small in comparison with the former. In fact, for $E_0 \sim 10^5$ esu, $d_0 \sim 10^{-18}$, $\Omega \sim 10^{15}$, $M \sim 10^{-24}$ and $c = 3 \times 10^{10}$, we find $q = 10^{-1}$ and $\mu = 10^{-10}$.

As will be shown below, the effectiveness of the acceleration of a dipole in an inhomogeneous RF field can be significantly increased by placing the particle in the high-frequency potential well formed by the field of the standing wave $E_x = E_0 \sin\omega t \sin(\omega z/c)$. In this case, substituting

$$d_x = d_0[a(t)\sin\omega t + b(t)\cos\omega t] \qquad (4)$$

into Eqs. (1) and (2), and averaging over the period of the RF field, we obtain

$$\dot{a} = \Delta b, \quad \dot{b} + \Delta a = -q(1 - a^2 - b^2)^{1/2}\sin\xi, \quad \ddot{\xi} = {}^1/_2\mu a\cos\xi, \qquad (5)$$

where we have introduced the notation

$$\tau = \omega t, \quad \xi = \frac{\omega z}{c}, \quad \Delta = 1 - \frac{\Omega}{\omega}, \quad q = \frac{d_0E_0}{\hbar\Omega}, \quad \mu = \frac{d_0E_0}{Mc^2}.$$

An interesting fact, following from (5), is that change in the amplitude of the dipole moment is associated with a displacement of the particle along the ξ-axis; if at the initial moment of time the molecule is at the bottom of the "well" (i.e., at the point $\xi = 0$, where the electric field vanishes) and is unpolarized; $a(0) = b(0) = 0$, a small displacement along the ξ-axis leads to polarization of the molecule, and this leads in turn to an increase in the shift, and so on. The beginning of the process ($a \ll 1$, $b \ll 1$, $\xi \ll 1$) can be investigated by linearizing Eq. (5). As a result we find the following equation for ξ:

*$[\mathbf{d}\,\mathrm{rot}\,\mathbf{E}] \equiv \mathbf{d} \times \mathrm{curl}\,\mathbf{E}$.

$$\xi^{(4)} + \Delta^2\ddot{\xi} + {}^1/_2\Delta\mu q\xi = 0. \quad (6)$$

Substituting $\xi = \xi_0 e^{\lambda\tau}$ in (6), we find

$$\lambda^2 = -{}^1/_2\Delta^2 \pm ({}^1/_4\Delta^4 - {}^1/_2\Delta\mu q)^{1/2}. \quad (7)$$

We are interested in the roots of Eq. (7) which have a positive real part; these correspond to the increase in the energy of the molecule. These roots occur for both signs of the frequency detuning. However, for $\Delta < 0$ increase in the energy of the oscillator occurs whatever the relation between the parameters μq and Δ, whereas for $\Delta > 0$ there is a threshold value $\Delta_c = (2\mu q)^{1/3}$. For $\Delta \lesssim \Delta_c$, Eq. (7) has two roots with a positive real part:

$$\lambda = \left(\frac{1}{8}\mu q\Delta\right)^{1/4}\left\{\left[1-\left(\frac{\Delta}{\Delta_c}\right)^{3/2}\right]^{1/2} \pm i\left[1+\left(\frac{\Delta}{\Delta_c}\right)^{3/2}\right]^{1/2}\right\} \quad (8)$$

From formulas (7) and (8) we can determine the maximum value of the increment $\gamma = \mathrm{Re}\,\lambda$, depending on Δ:

$$\gamma_{max} = ({}^1/_2\mu q)^{1/3}, \quad \Delta_{max} = -({}^1/_2\mu q)^{1/3},$$
$$\gamma_{max} = {}^1/_4\sqrt{3}(\mu q)^{1/3}, \quad \Delta_{max} = {}^1/_2(\mu q)^{1/3}. \quad (9)$$

To find the maximum energy acquired by the oscillator, it is necessary to examine the system of Eqs. (5) in a nonlinear approximation. In the general case, when the relationship between the parameters μ, q, and Δ is arbitrary, a solution can be found by means of numerical integration. Below we give analytic solutions of this system, which can be obtained if the conditions $\gamma \ll \gamma_{max}$ are fulfilled.

In the range of frequencies less than the resonance frequency ($\Delta < 0$), assuming that the condition $\dot{a} \ll \Delta a$ is fulfilled, expressing a in terms of ξ from the second of Eqs. (5):

$$a = \frac{q}{|\Delta|}\sin\xi\left(1+\frac{q^2}{\Delta^2}\sin^2\xi\right)^{-1/2} \quad (10)$$

and substituting this quantity into the equation of motion, we obtain

$$\ddot{\xi} = \frac{1}{2}\frac{q\mu}{|\Delta|}\sin\xi\cos\xi\left(1+\frac{q^2}{\Delta^2}\sin^2\xi\right)^{-1/2}. \quad (11)$$

The first integral of this equation

$$\dot{\xi}^2 = \frac{\mu|\Delta|}{q}\left[\left(1+\frac{q^2}{\Delta^2}\sin^2\xi\right)^{1/2} - \left(1+\frac{q^2}{\Delta^2}\sin^2\xi_0\right)^{1/2}\right] \quad (12)$$

allows us to determine the maximum displacement of the molecule. Substituting $\dot{\xi}(\xi_{max}) = 0$ in (12), we find $\xi_{max} = \pi + \xi_0$. If the condition $q^2 \ll \Delta^2$ is fulfilled, expanding the righthand side of (12) in a series and integrating, we find

$$\dot{\xi} = \frac{2\gamma\,\mathrm{tg}(\xi_0/2)\,e^{\gamma\tau}}{1+\mathrm{tg}^2(\xi_0/2)\,e^{2\gamma\tau}}, \quad a = \frac{q}{\gamma|\Delta|}\dot{\xi}, \quad \gamma = \left(\frac{1}{2}\frac{\mu q}{|\Delta|}\right)^{1/2}. \quad (13)$$

According to (13), the velocity of the particle increases exponentially at the beginning of the process (at $\xi \sim \xi_0$) and attains its maximum value $\dot{\xi}_{max} = \gamma$ when the particle is displaced to the maximum of the electric field $\xi = \pi/2$ (in a time $\tau_{max} = -\gamma^{-1}\ln\tan(\xi_0/2)$. The amplitude of the polarization vector attains its maximum value $a_{max} = q/|\Delta| \ll 1$ at this time).[1)]

After this the molecule moves into a retarding field and begins to lose energy.

The nonlinear approximation under consideration is correct provided that $(\mu q)^{1/3} \ll |\Delta|$, i.e., when the increment is small in comparison with its maximum. For an estimate we take $\gamma \sim \gamma_{max} = ({}^1/_2\,\mu q)^{1/3}$ and go over to dimensional variables; we find

$$v_{max} \approx c\left(\frac{1}{2}\frac{d_0^2E_0^2}{Mc^2\hbar\Omega}\right)^{1/3}. \quad (14)$$

Using the values given above for the parameters occurring in (14), we obtain $v_{max} \sim 10^7$ cm/sec.

We shall now consider the range of frequencies greater than the resonance frequency of the oscillator: $\Delta > 0$. In this case, using the relation

$$(\Delta^2 - \Delta^2a^2 - \dot{a}^2)^{1/2} = \Delta + qa\sin\xi - q\mu^{-1}\dot{\xi}^2, \quad (15)$$

it is possible to transform the system (5) to the following form:

$$\frac{d^2}{d\tau^2}\frac{\ddot{\xi}}{\cos\xi} + \Delta^2\frac{\ddot{\xi}}{\cos\xi} = -\frac{1}{2}q[\Delta\mu + q(2\dot{\xi}\,\mathrm{tg}\,\xi - \dot{\xi}^2)]\sin\xi. \quad (16)$$

An analytic solution of Eq. (16) can be obtained close to the instability threshold, i.e., for $\Delta \lesssim \Delta_c = (2\,\mu q)^{1/3}$, where the increment is small compared with Δ_c. A solution of (16) in this case can be sought in the form $\xi = \xi_0(\tau)\cos(\Delta\tau/\sqrt{2})$[5]. Substituting ξ into (16) and averaging the resulting expressions over the period Δ_c^{-1}, we find:[2)]

$$\ddot{\xi}_0 - \gamma^2\xi_0 = -\frac{1}{8}\left(\frac{\Delta_c^2}{3} + 3q^2\right)\xi_0^3, \gamma^2 = \frac{\Delta_c^2}{8}\left(\frac{\Delta_c^3}{\Delta^3} - 1\right) > 0. \quad (17)$$

Integrating (17), we determine the maximum displacement of the particle:

$$\xi_{0\,max} = 4\sqrt{3}\gamma(\Delta_c^2 + 9q^2)^{-1/2}. \quad (18)$$

Assuming that $\Delta_c^2 \ll q^2$ and taking $\gamma_{max} \sim (\mu q)^{1/3}$ for the estimate, we find $\xi_{max} \sim q^{-1}(\mu q)^{1/3}$.

Thus, if the principal effect in the field of a traveling wave is an increase in the internal energy of the molecule ($\xi_{max} \sim \mu$), in the field of a standing wave an increase in the translational energy of the molecule occurs, the conditions for acceleration being more effective in the region of frequencies $\omega < \Omega$ ($\dot{\xi}_{max} \sim \mu^{1/3}$ in comparison with $\dot{\xi}_{max} \sim \mu^{2/3}$ for $\omega > \Omega$).

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[1)] We should note that even at lower values of the frequency detuning $|\Delta| \sim q$, the quantity a, according to (10), cannot exceed unity, i.e., the molecule cannot go over into an inverted state.

[2)] Taking into account that $\xi_{0\,max} \ll 1$, we have expanded the trigonometric functions in (16) in series and have discarded terms of order higher than ξ_0^3.

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