Spontaneous radiative alignment of dipole moments of atoms moving in a medium

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The response of an oscillator moving in a medium to that radiation-force component which does no work but induces rotation is considered. The radiation torque is shown to rotate the dipole towards the direction of its motion. The dipole executes small oscillations about this direction. The frequency of these oscillations is determined. The effect leads to spontaneous alignment of the dipole moments as they pass through the medium. The feasibility of observing the effect in experiment is discussed.

The well-known reaction force exerted by radiation on its source was initially discussed for motion in vacuum,[1,2] and later also for emitters moving in a medium.[2–4] The overwhelming majority of papers deal with that reaction-force component which performs work and determines thereby the radiation power.

Yet the reaction-force component perpendicular to the source velocity, while performing no work, does induce rotation. It will be shown here that the reaction-force torque causes spontaneous alignment of radiating oscillators in the direction of their motion, oscillating about this direction.[1)]

The initial analysis deals by way of example with an oscillator moving at nonrelativistic velocity in a medium having a dielectric constant $\varepsilon(\omega) = 1 - \omega_0^2/\omega(\omega + i\nu_0)$ (ω_0 is the plasma frequency and ν_0 is the collision frequency). This model of the medium is quite general, since it describes a collisionless plasma and a weakly ionized collisional gas ($\omega \ll \nu_0$). Also discussed is spontaneous self-polarization in a gas of unperturbed two-level molecules, where $\varepsilon(\omega) = 1 - \Omega_c^2/(\omega^2 - \Omega_1^2)$ (Ω_c is known as the cooperative frequency of the gas[6] and Ω_1 is the transition frequency in a two-level system). In the latter case the polarization is due to the action exerted on the oscillator by the polariton modes it excites. The effect we are describing differs in this respect from the alignment of fast molecules by pair collisions,[7] since the alignment is caused here by collisions with collective excitations of the medium (plasmons, polaritons, and others).

To investigate the effect in question, the Poisson equation for the electric-field potential is used to determine the total force acting on the oscillator in the medium. This force has two components, one collinear with the oscillations plane, and the other perpendicular to this plane. The collinear component of the total force gives rise to a change in the vibrational energy of the oscillator (particularly to excitation if wave radiation in the region of the anomalous Doppler effect predominates[2,8,9]). Excitation of an oscillator by its own field has been the subject of many investigations (see Ref. 2 and the reference therein), and will not be discussed here. At the same time, the orthogonal reaction-force component rotates the oscillator and causes it to nutate about its motion axis. Thus, both the oscillator excitation and the alignment effects have a common cause, viz., action produced by dipole-excited natural waves of the medium.

Since the radiation force is weak under ordinary conditions[2] (in the present formulation, the sufficient condition for its weakness is the inequality $\Omega_n \ll \Omega$, where Ω_n is the frequency of the field-induced nutation), the excitation and alignment effects can be treated in first-order approximation independently, with the radiation force determined under the assumption that the oscillator motion is given. We note here that this procedure is not merely permissible but is necessary in the solution of problems involved the action of radiation force (lest paradoxes be encountered, such as charges that are accelerated under the influence of the self-field charges; see Ref. 2 for details). Expressions are obtained in this approximation for the nutation frequencies and damping rate. The paper concludes with estimates that point to the feasibility of observing the effect.

We proceed now to formulate the problem. Assume an oscillator moving with velocity $\mathbf{v} \uparrow \mathbf{z}_0$ in a medium and oscillating simultaneously at a frequency Ω in the xz plane, so that the oscillation plane makes an angle χ with the z axis. Assuming nonrelativistic motion, we begin with the Poisson equation for the potential of an electric field:

$$\hat{\varepsilon}\Delta\varphi = -4\pi\rho_{ext}, \qquad (1)$$

where $\rho_{\text{ext}} = e\delta(y)\delta(x - b\sin\Omega t)\delta(z - vt - a\sin\Omega t)$ is the imposed charge density, ε the dielectric constant of the medium, $\tan\chi = b/a$, and the oscillator peak-to-peak swing is $d = (a^2 + b^2)^{1/2}$. The source is assumed to move in a narrow channel located between the planes $y = \pm\delta$. If the channel half-width δ is much smaller than all the characteristic wavelengths of the system considered, the field in the vacuum gap is uniform in y (Ref. 2). Using these conditions, taking the Fourier transform with respect to $\mathbf{r}$ and t (see Ref. 6), we obtain an expression for the electric field produced by the oscillator in the channel:

$$\mathbf{E} = -\frac{i}{4\pi^2}\int \frac{\mathbf{k}\rho(\omega,\mathbf{k})\exp[-i\omega t + ik_z z + ik_x x - \delta(k_x^2+k_z^2)^{1/2}]}{\varepsilon(\omega)(k_x^2+k_y^2)^{1/2}} \times dk_z\, dk_x\, d\omega. \qquad (2)$$

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Here

$$\rho(\omega, \mathbf{k}) = 2\pi e \sum_{n=-\infty}^{\infty} J_n(k_z a + k_x b)\,\delta(\omega - k_z v - n\Omega)$$

is the Fourier transform of the imposed charge density and J_n is a Bessel function. To obtain the force acting on the oscillating charge, we must put $z = vt + a\sin\Omega t$, $x = b\sin\Omega t$ in Eq. (2) and multiply the result by the charge e; this yields

$$\mathbf{F} = -\frac{ie^2}{2\pi} \times \sum_{m,n=-\infty}^{\infty} \int \frac{\delta(\omega - k_z v - n\Omega) J_n(k_x b + k_z a) J_m(k_x b + k_z a)}{\varepsilon(\omega)\,[k_x^2 + k_z^2]^{1/2}} \mathbf{k}$$

$$\times \exp\{i(m-n)\Omega t\} \exp\{-(k_x^2 + k_z^2)^{1/2}\delta\}\, d\omega\, dk_x\, dk_z. \quad (3)$$

The nonzero averaged moment $\mathbf{M} = [\mathbf{r} \times \mathbf{F}]$ produces a force component proportional to $\sin\Omega t$.[2] To separate components of this type we must put $m = n \pm 1$ in (3) and use for the Bessel function the recurrence formula

$$J_{n-1}(x) - J_{n+1}(x) = 2dJ_n(x)/dx.$$

As a result of these transformations, the expression for the force takes the form

$$\mathbf{F}_0 = -\frac{e^2 \sin\Omega t}{2\pi} \times \sum_{n=-\infty}^{\infty} \int \frac{\delta(\omega - k_z v - n\Omega) \exp[-\delta(k_x^2 + k_z^2)^{1/2}]\, dJ_n^2(z)/dz}{\varepsilon(\omega)\,(k_x^2 + k_z^2)^{1/2}} \times \mathbf{k}\, dk_x\, dk_z\, d\omega,$$

$$z = bk_x + ak_z. \quad (4)$$

The moment of the force $\mathbf{F}_0$ is designated $\mathbf{M} = (0, M, 0)$ and is equal to

$$M = -\frac{e^2 \sin^2\Omega t}{2\pi} \times \sum_{n=-\infty}^{\infty} \int \frac{(ak_x - bk_z) \exp[-\delta(k_x^2 + k_z^2)^{1/2}]\, dJ_n^2(z)/dz}{\varepsilon(k_z v + n\Omega)\,(k_z^2 + k_x^2)^{1/2}}\, dk_x\, dk_z. \quad (5)$$

With new integration variables $z = ak_z + bk_x$ and $\tau = ak_x - bk_z$, Eq. (5) takes the simpler form

$$M = -\frac{e^2}{4\pi d} \sum_{n=-\infty}^{\infty} \int I(z, n) \frac{d}{dz} J_n^2(z)\, dz,$$

$$I(z, n) = \int_{-\infty}^{\infty} \frac{\tau \exp[-\nu(z^2 + \tau^2)^{1/2}]}{\varepsilon(\omega^*)\,(z^2 + \tau^2)^{1/2}}\, d\tau, \quad (6)$$

$$\omega^* = \frac{az - b\tau}{d^2} v + n\Omega, \qquad \nu = \frac{\delta}{d}.$$

It can be seen from (6) that the moment of the force is determined by the Hermitian part of the dielectric constant, whereas the radiation force and the polarization loss are determined by the anti-Hermitian part of ε.[3,4] The difference is due essentially to the fact that the torque-producing component of F performs no work and does not affect the radiation-loss power. This makes the analysis somewhat more difficult since, for example in a transparent medium, the radiation-force torque is determined by principal-value integrals of the form $⨍ d\omega f(\omega)/\varepsilon(\omega)$.

It can also be seen from (6) that if the oscillator oscillates along ($b = 0$) or across ($a = 0$) its motion, the moment M vanishes. Thus, an oscillator with orientation angles $\chi = 0$ or $\pi/2$ is in equilibrium.

Let us test the stability of these equilibrium states. To this end we transform the integral I into

$$I = \frac{1}{\pi} \int_{-\infty}^{\infty} K_0[|z|(t^2 + \nu^2)^{1/2}]\, dt \int_{-\infty}^{\infty} \frac{\tau e^{it\tau}}{\varepsilon(\omega^*)}\, d\tau, \quad (7)$$

where K_n is a modified Bessel function. To derive (7) we have used the formula

$$\frac{2}{\pi} \int_0^{\infty} K_0[|z|(t^2 + \nu^2)^{1/2}] \cos t\tau\, dt = \frac{\exp[-\nu(\tau^2 + z^2)^{1/2}]}{(\tau^2 + z^2)^{1/2}}.$$

Relations (6) and (7) can be further simplied by specifying an actual model of the medium. We consider for the sake of argument a collision-dominated plasma with $\varepsilon = 1 - \omega_0^2/\omega(\omega + i\nu_0)$ (ω_0 is the plasma frequency and ν_0 the frequency of the collisions between the electrons and the heavy particles). This choice of the dielectric constant is general enough, since it describes collisionless plasma as well as an absorbing medium ($\omega_0 \ll \nu_0$), when $\varepsilon = 1 + i\sigma/\omega$ ($\sigma = \omega_0^2/\nu_0$). After simple calculations in (7) we arrive at the relation

$$I = \frac{2i\bar\omega_0^2}{(\bar\omega_0^2 - \bar\nu_0^2/4)^{1/2}} \int_0^{\infty} K_1[|z|(t^2 + \nu^2)^{1/2}] \frac{t|z|}{(t^2 + \nu^2)^{1/2}} \times \exp\left\{it\left(\bar z + n\bar\Omega + \frac{i\bar\nu_0}{2}\right)\right\} \times \sin\left[\left(\bar\omega_0^2 - \frac{\bar\nu_0^2}{4}\right)^{1/2} t\right] dt, \qquad b > 0, \quad (8)$$

where $\bar z = az/b$, $\bar\Omega = \Omega d^2/bv$, $\bar\nu_0 = \nu_0 d^2/bv$, $\bar\omega_0 = \omega_0 d^2/bv$.

Substitution of (8) in (6) leads to a rather unwieldy equation that can, however, be further simplified. The simplification is made possible by the fact that the series

$$S = \sum_{n=-\infty}^{\infty} J_n^2(z) \exp(in\bar\Omega t) = J_0^2(z) + 2\sum_{n=1}^{\infty} J_n^2(z) \cos n\bar\Omega t$$

can be summed. Indeed, using the relation

$$J_n^2(z) = \frac{2}{\pi} \int_0^{\pi/2} J_{2n}(2z\cos\varphi)\, d\varphi,$$

we rewrite S in the form

$$S = \frac{2}{\pi} \int_0^{\pi/2} d\varphi \left[J_0(2z\cos\varphi) + 2\sum_{n=1}^{\infty} J_{2n}(2z\cos\varphi) \cos n\bar\Omega t\right]$$

$$= \frac{2}{\pi} \int_0^{\pi/2} d\varphi \cos\left(2z\cos\varphi \sin\frac{\bar\Omega t}{2}\right) = J_0\left(2z\sin\frac{\bar\Omega t}{2}\right).$$

As a result, Eq. (6) takes the form

$$\bar{M}=-\frac{2e^2\bar{\omega}_0^2}{\pi[\bar{\omega}_0^2-\bar{\nu}_0^2/4]^{1/2}d}$$

$$\times\int_0^\infty \frac{t\exp(-\bar{\nu}_0 t/2)\sin(\bar{\Omega}t/2)\sin[(\bar{\omega}_0^2-\bar{\nu}_0^2/4)^{1/2}t]}{(t^2+\nu^2)^{1/2}}dt$$

$$\times\int_0^\infty zK_1(\gamma z)J_1(\beta z)\sin\bar{z}t\,dz, \qquad (9)$$

where

$$\gamma=(t^2+\nu^2)^{1/2}, \quad \beta=2\sin(\bar{\Omega}t/2).$$

The integral with respect to z is expressed in terms of the spherical harmonic $Q_{1/2}(u)$. When the dipole approximation is applicable ($\Omega d/v\ll 1$), however, or when either $\tau\gg\beta$ or $at/b\gg\beta$, it suffices to calculate this integral approximately, with only the first term of the series expansion of the Bessel function. Using the tabulated integral

$$\int_0^\infty K_1(\gamma z)\sin\bar{z}t\,dz=\frac{\pi at}{2b(a^2t^2/b^2+\gamma^2)^{1/2}\gamma}$$

we rewrite (9) in the form

$$\bar{M}=-\frac{3\bar{\omega}_0^2e^2a}{bd(\bar{\omega}_0^2-\bar{\nu}_0^2/4)^{1/2}}$$

$$\times\int_0^\infty\frac{t^2\exp(-\bar{\nu}_0t/2)\sin^2(\bar{\Omega}t/2)\sin[(\bar{\omega}_0^2-\bar{\nu}_0^2/4)^{1/2}t]}{[(1+a^2/b^2)t^2+\nu^2]^{5/2}}dt. \qquad (10)$$

In the case of a collisionless plasma, the expression for the torque becomes

$$\bar{M}_1=-\frac{3e^2\bar{\omega}_0a}{bd}\int_0^\infty\frac{t^2\sin^2(\bar{\Omega}t/2)\sin\bar{\omega}_0t}{[(1+a^2/b^2)t^2+\nu^2]^{5/2}}dt, \qquad (11)$$

while for a weakly ionized collisional ($\nu_0\gg\omega_0$) we have

$$\bar{M}_2=-\frac{3\bar{\sigma}e^2a}{bd}\int_0^\infty\frac{t^2\exp(-\bar{\sigma}t)\sin^2(\bar{\Omega}t/2)}{[(1+a^2/b^2)t^2+\nu^2]^{5/2}}dt, \quad \bar{\sigma}=\frac{\sigma d^2}{bv}. \qquad (12)$$

Equation (11) remains finite also as $\nu\to0$. In this approximation we have

$$\bar{M}_1=-3\pi e^2\omega_0\Omega^2ab/8v^3. \qquad (13)$$

Equation (12) can be readily simplified if the inequalities $\sigma\ll\Omega, v/\delta\Omega\gg1$ hold. Then

$$\bar{M}_2=-(3e^2\sigma\Omega^2ab/4v^3)\ln(v/\delta\Omega). \qquad (14)$$

Similar results are obtained for the torque of the radiation force also if the oscillator moves in a gas of unexcited two-level molecules. Neglecting collisions, such a medium is described by a dielectric constant $\varepsilon=1-\Omega_c^2/(\omega^2-\Omega_1^2)$, where $\Omega_c^2=8\pi e^2d^2N\Omega_1/3\hbar$, N is the density of the molecules, and Ω_1 is the transition frequency. Omitting the intermediate calculations (which are similar to those given above), we write only the final result:

$$\bar{M}_3=-3\pi e^2\Omega_c^2\Omega^2ab/[\Omega_1^2+\Omega_c^2]^{1/2}v^3. \qquad (15)$$

Relations (13) and (14) show that the equilibrium state $\chi=\pi/2$ is unstable, but the equilibrium state $\chi=0$ is stable. Oscillators with initial random orientation become therefore aligned in the direction of their motion, oscillating about the equilibrium position $\chi=0$. Let us estimate the frequency of this oscillatory motion. Starting from the torque equation $Jd^2\chi/dt^2=M$, we obtain for motion in a collisionless plasma

$$\omega_1^2=3\pi e^2\omega_0\Omega^2/4mv^3, \qquad (16)$$

and for a weakly ionized gas with collisions

$$\omega_2^2=(3e^2\sigma\Omega^2/2mv^3)\ln(v/\delta\Omega). \qquad (17)$$

It is interesting to note that the nutation frequencies ω_1 and ω_2 do not depend on the dipole moment, but are determined by the parameters ω_0 and σ of the medium, by the natural frequency Ω, and by the particle velocity.

The nutating dipole emits electromagnetic waves at frequencies $\Omega\pm\omega_1$ or $\Omega\pm\omega_2$. Emission of the electromagnetic waves leads to damping of the oscillations. To estimate the damping rate, the torque of the radiation force[3)] $\mathbf{f}_p=(2e^2/3c^3)\dddot{\mathbf{r}}$, must be taken into account in the torque equation. Simple transformation recast the nutation equation in the form

$$\ddot{\chi}+2\dot{\chi}e^2\Omega^2/mc^3+\omega_{1,2}^2\chi=0, \qquad (18)$$

from which we see that the radiative damping rate of these oscillations is

$$\gamma_0=e^2\Omega^2/mc^3. \qquad (19)$$

We now estimate the characteristic frequencies of the nutation produced when fast ions move in a metal. Let an N^{6+} ion be channeled in a gold film along the [100] axis with velocity $v=2.4\cdot10^9$ cm/sec ($E\sim44$ MeV) and let a transition from level $n=1$ to level $n=2$ take place in the course of the motion.[10] Such transitions are due to resonant excitation of the electron spectrum by the crystal field.[10] The transition is then accompanied by torque-producing plasmon radiation. This radiation causes nutation at a relative frequency [see (16)]

$$\omega_1/\Omega=(3\pi e^2\omega_0/4mv^3)^{1/2}\approx2\cdot10^{-2}.$$

It seems likely that the effect considered in this paper can explain the experimentally observed splitting of the energy levels of ions traveling in metal films.[10] The experimentally obtained relative splittings $\Delta\omega/\omega\sim10^{-2}$ (Ref. 1) agree well with those calculated from Eq. (16).

The ions or atoms leaving the layer of matter become aligned, as already noted, predominantly in the direction of their motion. The self-polarization time is determined by Eq. (19). To estimate the polarization time we consider the transition $n=1\to n=2$ in the nitrogen ion N^{6+} (the frequency of this transition is $\Omega\sim5\cdot10^{17}\,\text{sec}^{-1}$). Using (19), we obtain $\gamma_0\sim0.4\cdot10^{13}\,\text{sec}^{-1}$, a polarization time $\tau_p\sim2\cdot10^{-13}$ sec, and a self-polarization length $L_p\approx5\cdot10^{-4}$ cm.

Self-polarization can be detected by measuring the absorption of polarized radiation that passes through a layer of ions. If the absorption increases when the polarization vector coincides with the direction of motion, this attests to spontaneous alignment of the dipole moments.

In conclusions, I wish to thank V. V. Kocharovskiĭ and V. Vl. Kocharovskiĭ for a helpful discussion of the results.

1)This effect is similar in some respects to radiative self-polarization of spins.[5] An essential difference, however, is the presence, in the case of radiative self-polarization of spins, of a longitudinal magnetic field along which the magnetic moments become aligned, whereas in our case there is no such field.

2)This is precisely why the presence of charges moving with constant velocity (e.g., of a nucleus with a nearby oscillator) does not alter the value of M, since the field produced by these charges is time-invariant in the moving reference frame.

3)We use here the expression for the radiation force in vacuum. This is permissible if the inequalities $\Omega \gg \sigma, \omega_0$ hold. In this case the dielectric constant ε tends to unity.

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