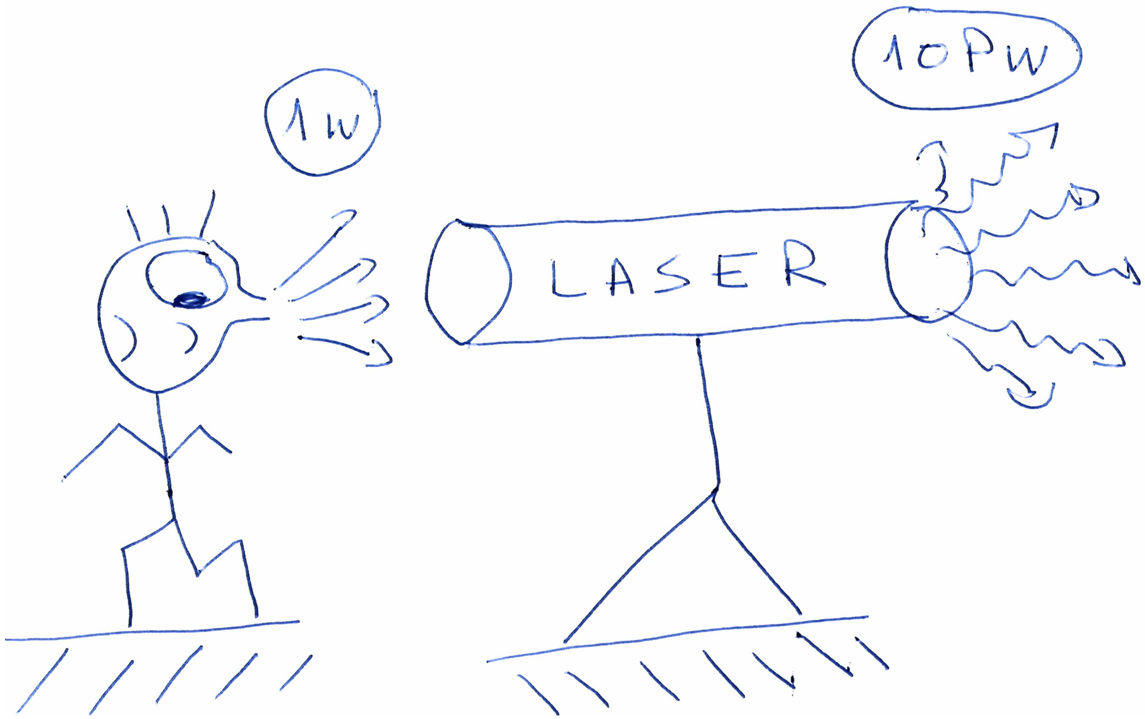


1W
10PW
LASER

ELI-NP at Magurele - "Pulse and Impulse of ELI"

1) "**Polaritonic pulse** and coherent X- and gamma rays from Compton (Thomson) backscattering" (MApostol&MGanciu), J. Appl. Phys. **109** 013307 (2011) (1-6)

2)"Dynamics of **electron–positron pairs** in a vacuum polarized by an external radiation field" (MA), Journal of Modern Optics, **58** 611 (2011)

3)"**Classical interaction** of the electromagnetic radiation with two-level polarizable matter" (MA), Optik **123** 193 (2012)

4)"**Coherent polarization** driven by external electromagnetic fields" (MA&MG), Physics Letters **A374** 4848 (2010)

5)"Coupling of **(ultra-) relativistic atomic nuclei** with photons" (MA&MG), AIP Advances **3** 112133 (2013)

6)"Propagation of **electromagnetic pulses** through the surface of dispersive bodies" (MA), Roum J. Phys. **58** 1298 (2013)

7)"**Giant dipole oscillations** and ionization of heavy atoms by intense electromagnetic pulses" (MA), Roum. Reps. Phys. (2015)

8)"**Parametric resonance**" in rotation molecular spectra" (MA)

INSTITUTE of PHYSICS and NUCLEAR ENGINEERING
Magurele-Bucharest

Parametric resonance in rotation molecular spectra

or

Rotation molecular spectra in static electric fields

M Apostol

April 2015

What can we do with (high-power) lasers and nuclei?

1) Lasers accelerate (plasma) electrons and ions (p); 10MeV, good flux

2) Electrons $\rightarrow\gamma$ (bremsstrahlung; Compton); 10MeV; good flux

3) Nuclear reactions: fission, (p,n)-emission, transmutation, n-sources

Improve nuclear data, applications (isotopes, transm)

Other, different, new Nuclear Phys

Lasers produce **strong and very strong** electric (magnetic) fields

Nuclei in strong fields: change of levels$\rightarrow$change in reaction rate, decay

(Lasers fields slow $\longleftrightarrow$ nuclear processes)

Very similar with Molecules in Strong Fields

With a difference: Laser fields are fast $\longleftrightarrow$ molecular processes

Strong time-dependent electric fields: $E_0 \cos \Omega t$, $\Omega = 2\pi \cdot 10^{15} s^{-1}$ ($1 eV$)

$10^{20} w/m^2 \to E_0 = 10^9 statvolt/cm$ (compare at fields 10^6)

(Not as high as Schwinger limit 10^{13} and non-linear QED!)

Accel $\frac{qE_0}{m}$, velocity $\frac{qE_0}{m\Omega}$, path $d = \frac{qE_0}{m\Omega^2}$, compare with l (atoms, mols, nuclei)

Nuclei: $d = 10^{-8} cm \gg l$: shift of en levels

Mols: similar, $d = 10^{-8} cm \sim l$; on the border

Atoms: $d = 0.1\mu \gg l$: shift the levels

What happens:

$\mathcal{E}_m \to \mathcal{E}_m + \frac{qcE_0}{\Omega} \cos \Omega t$

Dressed states: $e^{-\frac{i}{\hbar}(\mathcal{E}_m + n\Omega)t}$ (coherent states)

Transitions, decay, etc

Molecular Phys in Strong Fields

(ionization, dissociation, chem reactions)

Molecular spectroscopy

First, in static el fields (then in fast el fields)

Generalities (well known)

Molecules, el dipole moment $d = 10^{-18} esu$

Spherical pendulum (spherical top, spatial, rigid rotator)

Coupling time-dependent el field $\Longrightarrow$ (free) rotation (and vibration) spectra

$\nu = 10^{11} - 10^{13} s^{-1}$ (infrared)

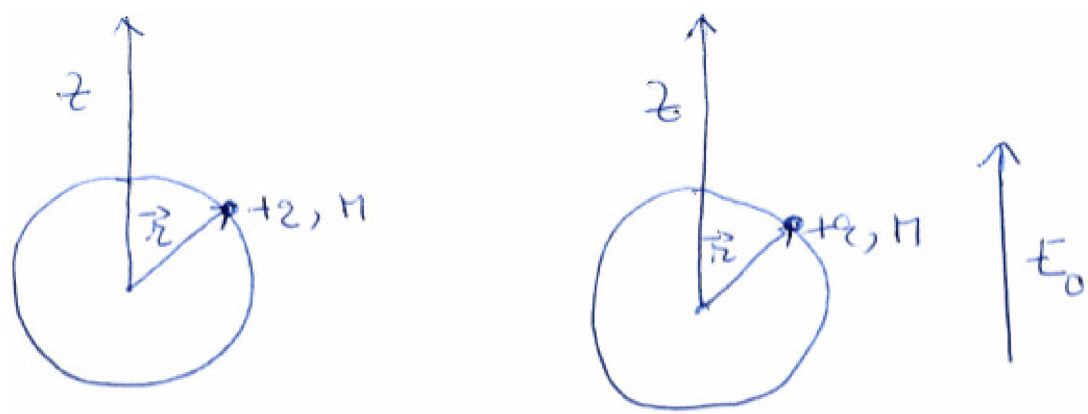
Special situations (less known)

External static electric field (highly-oscillating fields?)

Internal static electric field (polar matter; pyroelectrics, ferro-electrics)

Low temperatures

Heavy polar impurities



Free rotations: Approx azimuthal rotations+zenithal oscillations

$$H = \frac{1}{2}M\dot{\mathbf{l}}^2 = \frac{1}{2}Ml^2(\dot{\theta}^2 + \dot{\varphi}^2 \sin^2\theta) = \frac{L^2}{2I}$$

$L_x = Mr^2(-\dot{\theta}\sin\varphi - \dot{\varphi}\sin\theta\cos\theta\cos\varphi), L_y = Mr^2(\dot{\theta}\cos\varphi - \dot{\varphi}\sin\theta\cos\theta\sin\varphi$
$L_z = Mr^2\dot{\varphi}\sin^2\theta$

$$L^2 = -\hbar^2\left[\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}(\sin\theta\frac{\partial}{\partial\theta}) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}\right]$$

Y_{lm}, $\hbar^2 l(l+1)$, $l = 0, 1, ...;$ $L_z = -i\hbar\frac{\partial}{\partial\varphi}$, $L_z Y_{lm} = \hbar m Y_{lm}$, $m = -l, -l+1, ...l.$; degeneracy $2l+1$

Classical eqs of motion

$$\ddot{\theta} = \dot{\varphi}^2 \sin\theta \cos\theta \ , \ I\frac{d}{dt}(\dot{\varphi}\sin^2\theta) = 0$$

$\dot{\varphi} = L_z/I\sin^2\theta$; conserved $\mathbf{L}$

$$H = \frac{1}{2}I\dot{\theta}^2 + \frac{L_z^2}{2I\sin^2\theta}$$

Effective potential function $U_{eff} = L_z^2/2I\sin^2\theta$, minimum for $\theta = \pi/2$,$\delta\vartheta = \theta - \pi/2$

$$H \simeq \frac{1}{2}I\delta\dot{\theta}^2 + \frac{L_z^2}{2I}\delta\theta^2 + \frac{L_z^2}{2I}$$

Precession $\varphi = \omega_0 t$, $\omega_0 = L_z/I$, oscillation $\delta\theta = A\cos(\omega_0 t + \delta)$

Coupling: $H_{int}(t) = -dE \cos\theta \cos\omega t$

Eqs

$$\ddot{\theta} = \dot{\varphi}^2 \sin\theta \cos\theta - \frac{dE}{I} \sin\theta \cos\omega t \ ,$$

$$I \frac{d}{dt}(\dot{\varphi} \sin^2\theta) = 0 \ ;$$

$U_{eff} = \frac{L_z^2}{2I \sin^2\theta}$

Harmonic-oscillator

$$\delta\ddot{\theta} + \omega_0^2 \delta\theta = -\frac{dE}{I} \cos\omega t$$

where $\omega_0 = L_z/I = \hbar m/I$

Solution

$$\delta\theta = a\cos\omega t + b\sin\omega t$$

$$a = \tfrac{dE}{2I\omega_0}\tfrac{\omega-\omega_0}{(\omega-\omega_0)^2+\gamma^2}\ ,\ \ b = -\tfrac{dE}{2I\omega_0}\tfrac{\gamma}{(\omega-\omega_0)^2+\gamma^2}$$

typical resonance

Approx: $L_z \simeq L$ ($m \simeq l$, $L_x^2 + L_y^2 \ll L_z^2 \simeq L^2$)

Mean absorbed power

$$P = -dE\overline{\delta\dot{\theta}\cos\omega t} = -\frac{1}{2}dEb\omega_0 = \frac{d^2E^2}{4I}\frac{\gamma}{(\omega-\omega_0)^2+\gamma^2}$$

QM (exact)

$$\omega_0 = (E_{l+1} - E_l)/\hbar = (\hbar/I)(l+1)$$

$$\frac{\partial\,|c_{lm}|^2}{\partial t} = \frac{\pi d^2 E^2}{2\hbar^2}\,|(\cos\theta)_{lm}|^2\,\delta(\omega_0 - \omega)$$

$$(\cos\theta)_{lm} = (\cos\theta)_{l+1,m;l,m} = -i\sqrt{\frac{(l+1)^2 - m^2}{(2l+1)(2l+3)}}$$

$$P_q = \hbar\omega_0 \textstyle\sum_{m=-l}^{l} \frac{\partial |c_{lm}|^2}{\partial t} = \frac{\pi d^2 E^2}{2\hbar}\omega_0 \sum_{m=-l}^{l} |(\cos\theta)_{lm}|^2\,\delta(\omega_0 - \omega) =$$

$$= \tfrac{d^2E^2}{6\hbar}\omega_0(l+1)\tfrac{\gamma}{(\omega-\omega_0)^2+\gamma^2} = \tfrac{d^2E^2}{6I}(l+1)^2\tfrac{\gamma}{(\omega-\omega_0)^2+\gamma^2}$$

Finite temperatures

$$P_{q,th} = \tfrac{\pi d^2 E^2}{2\hbar}\omega_0 \times$$

$$\times \textstyle\sum_{m=-l}^{l} |(\cos\theta)_{lm}|^2 \left[e^{-\beta\hbar^2 l(l+1)/2I} - e^{-\beta\hbar^2 (l+1)(l+2)/2I}\right] \delta(\omega_0 - \omega)/Z$$

$$Z = \sum_{l=0} (2l+1) e^{-\beta\hbar^2 l(l+1)/2I} = \frac{2I}{\beta\hbar^2}$$

is the partition function

$$P_{q,th} = \tfrac{\pi d^2 E^2}{12I}(l+1)^3 \left(\tfrac{\beta\hbar^2}{I}\right)^2 e^{-\beta\hbar^2 l(l+1)/2I} \delta(\omega_0 - \omega) =$$

$$= \tfrac{1}{2} P_q (l+1) \left(\tfrac{\beta\hbar^2}{I}\right)^2 e^{-\beta\hbar^2 l(l+1)/2I}$$

Typical values: $I = 10^{-38} g \cdot cm^2$ (molecular mass $M = 10^5$ electronic mass $m = 10^{-27} g$, the dipole length $r = 10^{-8} cm$ (1Å)), and get $\hbar / I = 10^{11} s^{-1} \simeq 1K$ ($\omega_0 = \hbar m / I$, or $\omega_0 = \hbar(l+1)/I$)

Room temperature $\beta \hbar^2 (l+1)/I \ll 1$) (many levels)

Harmonic oscillator, energy levels $\hbar\omega_0(n+1/2)$, $n = 0,1,2...$, $\omega_0 = L_z/I = \hbar m/I$, $m = 0,1,2...$; $\omega_0 = \hbar m/I \rightarrow$q-m frequency $\omega_0 = (E_{l+1} - E_l)/\hbar = (\hbar/I)(l+1)$

Transitions $n \to n+1$, absorbed power

$$P_n = \frac{\pi d^2 E^2}{4I}(n+1)\delta(\omega_0 - \omega)$$

Total power

$$P_{osc} = \sum_{n=0}^{N} P_n = \frac{\pi d^2 E^2}{2I} m(m+1/2)\delta(\omega_0 - \omega)$$

$$(\delta\theta)_{N+1,N} = \sqrt{\frac{\hbar(N+1)}{2I\omega_0}} = \sqrt{\frac{N+1}{2m}} \ll 1$$

Compares well with the exact q-m result - h-osc satisfactory approx

Strong Static El Field

$$H = \frac{1}{2} I (\dot{\theta}^2 + \dot{\varphi}^2 \sin^2 \theta) - dE_0 \cos\theta$$

Cons of L_z

$$I \frac{d}{dt} (\dot{\varphi} \sin^2 \theta) = 0$$

Effective potential function

$$U_{eff} = \frac{L_z^2}{2I \sin^2 \theta} - dE_0 \cos\theta$$

Assume: $dE_0 \gg L_z^2/I \sim T \Longrightarrow E_0 \gg T/d = 4 \times 10^4 esu\,(1.2 \times 10^9 V/m)$

Very high; atomic fields $4.8 \times 10^6 esu$

Polar matter (*e.g.*, pyroelectrics, ferroelectrics), OK!

Low temperatures, free molecular rotations hindered

dipoles quenched, execute small rotations and vibrations

Transitions from free rotations to small vibrations around quenched positions in polar matter is seen in the curve of the heat capacity *vs* temperature (Pauling, 1930)

Similarly, sstrong static electric fields may appear locally near polar impurities with large moments of inertia, embedded in polar matter.

U_{eff} minimum, for $\theta_0 \simeq (L_z^2/IdE_0)^{1/4} \simeq (T/dE_0)^{1/4} \ll 1$

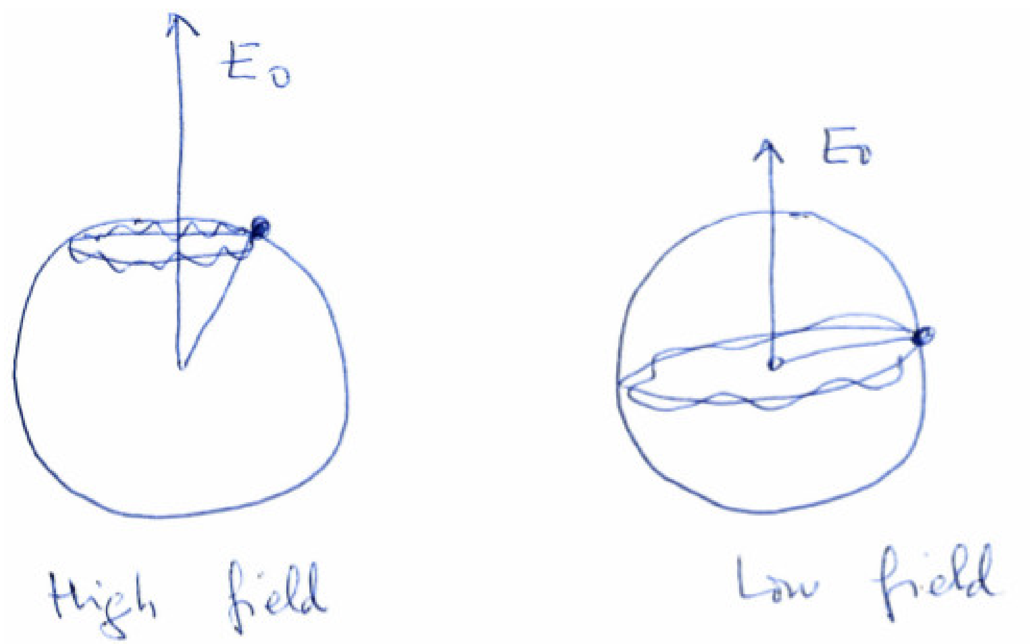
Harmonic oscillator

$$U_{eff} \simeq -dE_0 + 2dE_0\delta\theta^2$$

$$H \simeq \frac{1}{2}I\delta\dot{\theta}^2 + \frac{1}{2}I\omega_0^2\delta\theta^2 - dE_0$$

$\omega_0 = 2\sqrt{dE_0/I} \gg 10^{12}s^{-1}$ (Rabi's frequency, 1936)

Worth noting: frequency ω_0 given by the static field E_0


E0
E0
High field
Low field.

Coupling:

$$H_{int} = -dE(t)(\sin\alpha\sin\theta\cos\varphi + \cos\alpha\cos\theta)$$

$$H_{1int} = -\tfrac{1}{2}dE\sin\alpha\left[\cos(\omega + \tfrac{1}{2}\omega_0)t + \cos(\omega - \tfrac{1}{2}\omega_0)t\right]\delta\theta\ ,$$

$$H_{2int} = \tfrac{1}{2}dE\cos\alpha\cos\omega t \cdot \delta\theta^2\ .$$

H_{1int}: transitions $n \to n+1$, resonance frequency

Absorbed power

$$P_q = \tfrac{\pi}{16I\omega_0}d^2E^2\Omega(n+1)\sin^2\alpha\delta(\omega-\Omega) =$$

$$= \tfrac{1}{16I\omega_0}d^2E^2\Omega(n+1)\sin^2\alpha\tfrac{\gamma}{(\omega-\Omega)^2+\gamma^2}\ ,\ \gamma \to 0^+$$

(resonance), $\Omega = \frac{1}{2}\omega_0,\ \frac{3}{2}\omega_0$.

Temperature dependence

$$P_{q,th} = \frac{\pi}{16I\omega_0} d^2 E^2 \Omega \sum_{n=0} (n+1) \left[e^{-\beta\hbar\omega_0 n} - e^{-\beta\hbar\omega_0 (n+1)} \right] \times$$

$$\times \sin^2 \alpha \delta(\omega - \Omega) / \sum_{n=0} e^{-\beta\hbar\omega_0 n}$$

where the summation over n is, in principle, limited.

Validity: $(\delta\theta)_{n+1,n} = \sqrt{\hbar/2I\omega_0}\sqrt{n+1} \ll \theta_0 \simeq (L_z^2/IdE_0)^{1/4} \Longrightarrow$ $n \ll 80$ Extend the summation, $P_{q,th}$ independent of temperature

Validity: $L_x^2 + L_y^2 \simeq L^2 \gg L_z^2$.

Parametric resonance

$$H^{'} = H + H_{2int} = \frac{1}{2} I \delta\dot{\theta}^2 + \frac{1}{2} I \omega_0^2 (1 + h \cos \omega t) \delta\theta^2$$

($h = \frac{E}{2E_0} \cos \alpha$), Mathieu's eq

$$\delta\ddot{\theta} + \omega_0^2 (1 + h \cos \omega t) \delta\theta = 0$$

Periodic solutions, aperiodic solutions, which may grow indefinitely with increasing the time for ω near $2\omega_0/n$, $n = 1, 2, 3...$

Initial conditions, thermal fluctations, class sol ineffective

QM: different!

Transitions $n \to n+2$ (double quanta abs, Goeppert-Mayer, 1931)

Absorbed power

$$P_q = 2\hbar\omega_0 \frac{\partial \left| c_{n+2,n} \right|^2}{\partial t} = \frac{\pi h^2}{64} \hbar\omega_0^3 (n+1)(n+2)\delta(2\omega_0 - \omega) =$$

$$= \frac{h^2}{64} \hbar\omega_0^3 (n+1)(n+2) \frac{\gamma}{(2\omega_0 - \omega)^2 + \gamma^2}\ ,\ \gamma \to 0^+$$

$$P_{q,th} = \frac{\pi h^2}{64} \hbar\omega_0^3 \sum_{n=0} (n+1)(n+2) \times$$

$$\times \left[e^{-\beta\hbar\omega_0(2n+1)} - e^{-\beta\hbar\omega_0(2n+3)} \right] \delta(2\omega_0 - \omega) / \left[\sum_{n=0} e^{-\beta\hbar\omega_0 n} \right]^2$$

The parametric resonance disappears for $\alpha = \frac{\pi}{2}$ ($\mathbf{E}$ right angle $\mathbf{E}_0$)

Orientation of the solid ($\overline{\cos^2 \alpha} = \frac{1}{3}$)

Parameter γ small (dipolar interaction), sharp lines

Liquids, beside average over angle α, motional narrowing

Gases, the quenching field is weak, and the parametric resonance is not likely to occur

Weak Static El Field

E_0, $dE_0 \ll L_z^2/I \sim T$

Effective potential U_{eff} minimum for $\theta \simeq \frac{\pi}{2}$

Expansion $\widetilde{\theta} = \theta - \frac{\pi}{2}$, harmonic oscillator, $\omega_0 = L_z/I$

$$H \simeq \frac{1}{2} I \dot{\widetilde{\theta}}^2 + \frac{1}{2} I \omega_0^2 \dot{\widetilde{\theta}}^2$$

E_0 brings only a small correction to the $\pi/2$-shift in θ

Contribution to the hamiltonian is a second-order effect

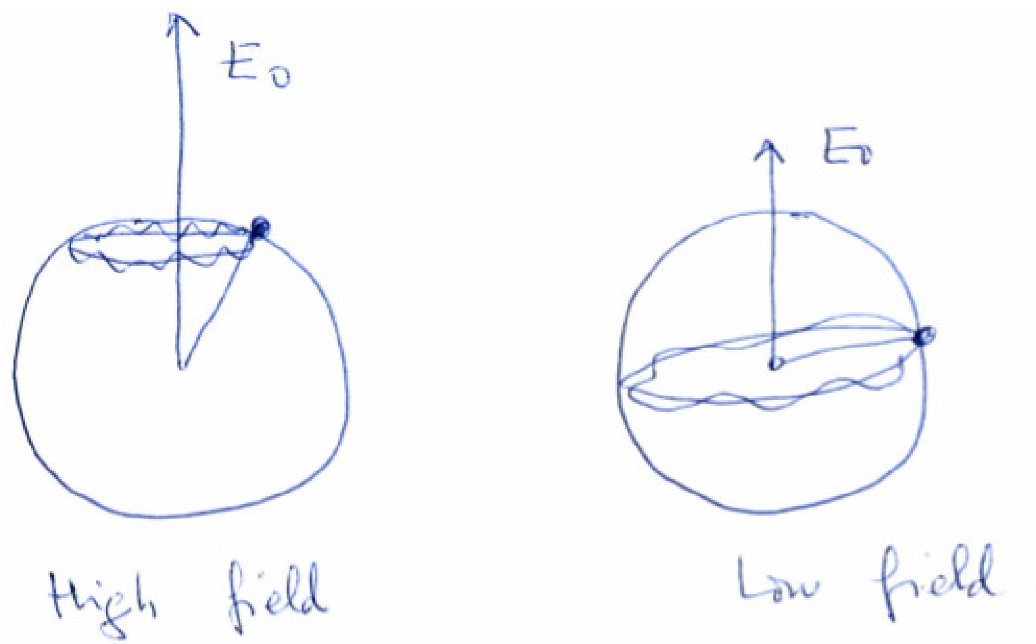
φ moves freely with $\dot{\varphi} = \omega_0$

In contrast with the high-field case (where the frequency $\dot{\varphi}$ is fixed by the static field E_0, in the low-field case we may quantize the φ-motion, according to $L_z = \hbar m$, m integer, such that $\omega_0 = \frac{\hbar}{I} m$

Lowest value is $\hbar / I \simeq 10^{11} s^{-1} (1K)$

The molecular rotations are described by a set of harmonic oscillators with frequencies $\omega_0 = \frac{\hbar}{I} m$, beside the φ-precession

Valid for $n \ll m$ (sufficiently large at room temprature, $m = 300$) $(L_x^2 + L_y^2 \ll L^2 \simeq L_z^2 \ (m \simeq l))$


E_0
E_0
High field
Low field.

Coupling

$$H_{1int} = dE \cos\alpha \cos\omega t \cdot \widetilde{\theta} \ ,$$

$$H_{2int} = \tfrac{1}{4} dE \sin\alpha \left[\cos(\omega + \omega_0)t + \cos(\omega - \omega_0)t\right] \cdot \widetilde{\theta}^2$$

H_{1int}: transitions $n \to n+1$, absorbed power

$$P_q = \frac{\pi}{4I} d^2 E^2 (n+1) \cos^2\alpha \delta(\omega_0 - \omega)$$

For n, m ($n \ll m$) sum over a few values of m in $\delta(\omega_0 - \omega) = \delta(\hbar m/I - \omega)$ with the statistical weight $e^{-\beta\hbar^2 m^2/2I}$

For $\hbar/I \gg \gamma$, a few, distinct absorption lines at $\omega_0 = \hbar m/I$ (band of absorption)

Temperature dependence

$$P_{q,th} = \frac{\pi}{4I} d^2 E^2 \cos^2\alpha \cdot C \sum_{m>0} e^{-\beta\hbar^2 m^2/2I} \times$$

$$\times \left\{ \sum_{n=0} (n+1) \left[e^{-\beta\hbar\omega_0 n} - e^{-\beta\hbar\omega_0 (n+1)} \right] / \sum_{n=0} e^{-\beta\hbar\omega_0 n} \right\} \delta(\omega_0 - \omega)$$

$\omega_0 = \hbar m/I$, $C \sum_{m>0} e^{-\beta\hbar^2 m^2/2I} = 1$

Envelope

$$P_{q,th} = \frac{\pi}{4} d^2 E^2 \cos^2\alpha \sqrt{\frac{2\pi\beta}{I}} e^{-\beta I\omega^2/2}$$

Interaction H_{2int}: transitions $n \to n+2$ (frequency $2\omega_0$) for external frequencies $\Omega = \omega_0$, $3\omega_0$ (superposed over the transitions produced by H_{1int})

$$P_q = \frac{\pi\hbar\Omega}{128 I^2 \omega_0^2} d^2 E^2 (n+1)(n+2) \sin^2\alpha \delta(\Omega - \omega)$$

$$P_{q,th} = \tfrac{\pi\hbar}{128 I^2} d^2 E^2 \sin^2\alpha \cdot C \textstyle\sum_{m>0} \tfrac{\Omega}{\omega_0^2} e^{-\beta\hbar^2 m^2/2I} \times$$

$$\times \left\{ \textstyle\sum_{n=0} (n+1)(n+2) \left[e^{-\beta\hbar\omega_0(2n+1)} - e^{-\beta\hbar\omega_0(2n+3)} \right] \right\} /$$

$$/ \left[\textstyle\sum_{n=0} e^{-\beta\hbar\omega_0 n} \right]^2 \delta(\Omega - \omega)$$

$$P_{q,th} = \frac{\pi\hbar}{64 I^2} d^2 E^2 \sin^2\alpha \cdot C \sum_{m>0} \frac{\Omega}{\omega_0^2} e^{-\beta\hbar^2 m^2/2I} \frac{e^{-\beta\hbar\omega_0}}{(1 + e^{-\beta\hbar\omega_0})^2} \delta(\Omega - \omega)$$

Note: the weak field E_0 does not appear explicitly in the above formulae

Its role: setting the z-axis, highlight the directional effect of E (angle α), reduce the conservation $\mathbf{L} \to L_z$

Comparison with free rotations: same frequencies $\omega_l = \hbar(l+1)/I$, $l = 0, 1, 2...$ as $\omega_0 = \hbar m/I$, $m = 0, 1, 2...$

Weak field: statistical behaviour:

$$H = \frac{1}{2} I (\dot{\theta}^2 + \dot{\varphi}^2 \sin^2 \theta)$$

or

$$H = \frac{1}{2I} P_\theta^2 + \frac{1}{2I \sin^2 \theta} P_\varphi^2$$

momenta $P_\theta = I\dot{\theta}$ and $P_\varphi = I\dot{\varphi} \sin^2 \theta$

Classical statistical distribution

$$const \cdot dP_\theta dP_\varphi d\theta e^{-\beta P_\theta^2/2I} e^{-\beta P_\varphi^2/2I \sin^2 \theta}$$

or, integrating over momenta, $\frac{1}{2} \sin \theta d\theta$

In the presence of the field, distribution $\simeq \frac{1}{2} \sin \theta d\theta \cdot e^{\beta \mathbf{d} \mathbf{E}_0}$ which leads to $\overline{\cos \theta} = \beta d E_0 / 3$ (Curie-Langevin-Debye law, 1895-1912)

QM: $dE_0 \ll \hbar^2/I$, interaction $-dE_0 \cos\theta$ brings a second-order contribution to the energy levels $E_l = \hbar^2 l(l+1)/2I$, there appear diagonal matrix elements of $(\widetilde{\cos\theta})_{lm,lm}$ in the first-order of the perturbation theory, and the mean value is given by $\overline{\cos\theta} = \sum(\widetilde{\cos\theta})_{lm,lm}\Delta(\beta E_l)e^{-\beta E_l}/\sum e^{-\beta E_l} = \beta dE_0/3$

Dipole interaction

Many molecules: el dipole d (in their gs)

Rarefied cond matter: el dipoles randomly distributed

Slightly aligned by an $\mathbf{E}_0$, leading induced orient pol $\overline{d} = \beta d^2 E_0/3$

(Curie-Langevin-Debye law)

Interaction: $d = 10^{-18} esu$, $a = 10^{-8} cm$ (1Å) $\Longrightarrow \simeq d^2/a^3 = 10^{-12} erg \simeq 10^3 K$

Not a small energy! (Field $d/a^3 = 10^6 statvolt/cm$)

Interaction energy

$$U = -\frac{3(\mathbf{d}_1\mathbf{d}_2)a^2 - (\mathbf{d}_1\mathbf{a})(\mathbf{d}_2\mathbf{a})}{a^5}$$

(θ_1, φ_1), (θ_2, φ_2) with respect to the axis $\mathbf{a}$

$$U = -\frac{d_1 d_2}{a^3}[2\cos\theta_1\cos\theta_2 + 3\sin\theta_1\sin\theta_2\cos(\varphi_1 - \varphi_2)]$$

Four extrema: $\theta_1 = \theta_2 = 0, \pi/2$ and $\varphi_1 - \varphi_2 = 0, \pi$

Only $\theta_1 = \theta_2 = \pi/2$, $\varphi_1 - \varphi_2 = 0$ is a local minimum

Near the minimum

$$U = \tfrac{d_1 d_2}{a^3}[-3 + \tfrac{3}{2}(\delta\theta_1^2 + \delta\theta_2^2) - 2\delta\theta_1\delta\theta_2 + \tfrac{3}{2}(\delta\varphi_1 - \delta\varphi_2)^2] =$$

$$= \tfrac{d_1 d_2}{a^3}[-3 + \tfrac{1}{4}(\delta\theta_1 + \delta\theta_2)^2 + \tfrac{5}{4}(\delta\theta_1 - \delta\theta_2)^2 + \tfrac{3}{2}(\delta\varphi_1 - \delta\varphi_2)^2]$$

where $\delta\theta_{1,2} = \theta_{1,2} - \pi/2$, $\delta\varphi_{1,2}$ $(\varphi_1 - \varphi_2 = 0)$

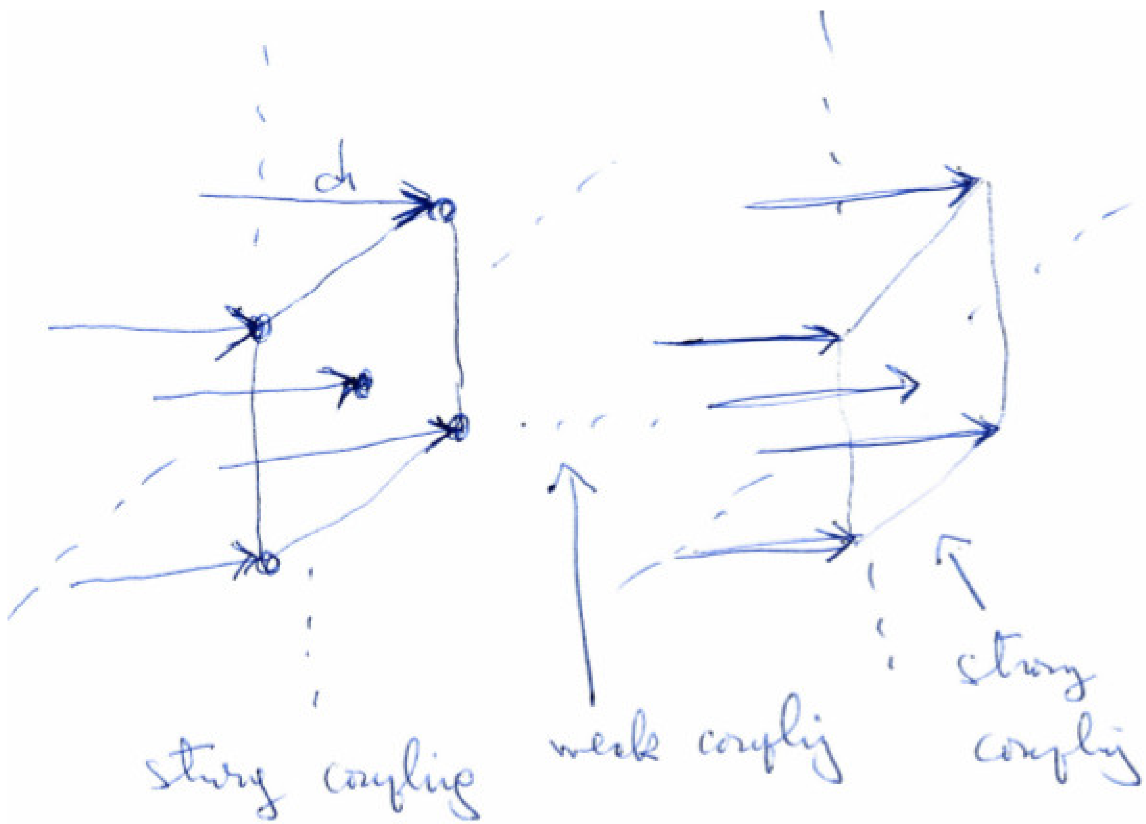
The el dipoles are **quenched** in equilibrium positions $\theta_1 = \theta_2 = \pi/2$, $\varphi_1 - \varphi_2 = 0$!

Small rotations and vibrations around

The dipoles are (spontaneously) aligned along an arbitrary axis

El (macroscopic) polarization

Substances with a permanent electric polarization: pyroelectrics (or electrets); ferroelectrics (paraelectrics) (piezoelectricity)


dh
strong coupling
weak coupling
strong
coupling

Example: barium titanate ($BaTiO_3$)

Elementary cell $a \simeq 4 \times 10^{-8} cm$ (4Å)

Dipole of a cell $d \simeq 5 \times 10^{-18} esu$

Displacement $\delta = 0.1$Å $\ll a$

Structural modifications (cubic to tetragonal to monoclinic to rhombohedral with decreasing temperature)

Elongation

Continuum model (possibly two-dimensional)

$$H_{int} = \frac{1}{a^3}\int d\mathbf{r}\left[\frac{d^2}{a^3}\delta\theta^2 + \frac{5d^2}{4a}(grad\delta\theta)^2 + \frac{3d^2}{2a}(grad\delta\varphi)^2\right]$$

Full hamiltonian

$$H = \frac{1}{a^3}\int d\mathbf{r}[\tfrac{1}{2}I\dot{\delta\theta}^2 + \tfrac{1}{2}I\dot{\delta\varphi}^2 + \tfrac{1}{2}I\omega_0^2\delta\theta^2 +$$

$$+\tfrac{1}{2}Iv_\theta^2(grad\delta\theta)^2 + \tfrac{1}{2}Iv_\varphi^2(grad\delta\varphi)^2$$

$\omega_0^2 = 2d^2/Ia^3$, $v_\theta^2 = 5d^2/2Ia = 5\omega_0^2a^2/4$, $v_\varphi^2 = 3d^2/Ia = 3\omega_0^2a^2/2$

Dipolar waves (waves of orientational polarizability), elementary excitations

Wave equations

$$\ddot{\delta\theta} + \omega_0^2\delta\theta - v_\theta^2\Delta\delta\theta = 0\ ,\ \ddot{\delta\varphi} - v_\varphi^2\Delta\delta\varphi = 0$$

Spectrum $\omega_\theta^2 = \omega_0^2 + v_\theta^2 k^2$, $\omega_\varphi^2 = v_\varphi^2 k^2$

$\omega_0 \simeq 10^{13} s^{-1}$ (infrared region), velocities $v_{\theta,\varphi} \simeq 10^5 cm/s$ (the wavelengths are $\lambda_{\theta,\varphi} \simeq \pi\sqrt{5}a$, $\pi\sqrt{6}a$)

Polar-matter modes: "**dipolons**"

Coupling: $\mathbf{E}(\mathbf{r},t) = \mathbf{E}\cos(\omega t - \mathbf{kr})$

$$H^{'} = -\frac{1}{a^3}\int d\mathbf{r}\mathbf{d}\mathbf{E}\cos(\omega t{-}\mathbf{kr})$$

$$H^{'} = -\frac{1}{a^3}\int d\mathbf{r}dE(\delta\theta\sin\alpha - \frac{1}{2}\delta\theta^2\cos\alpha)\cos(\omega t{-}\mathbf{kr})$$

φ-waves do not couple

Since the wavelength of the radiation field $\gg$the wavelength of the dipolar interaction ($v_{\theta,\varphi} \ll c$, where c is the speed of light), we may drop out the spatial dependence (spatial dispersion)

Equation of motion of a harmonic oscillator

$$\ddot{\delta\theta} + \omega_0^2\delta\theta = \frac{dE}{I}\sin\alpha\cos\omega t - \frac{dE}{I}\delta\theta\cos\alpha\cos\omega t$$

First interaction term gives

$$\delta\ddot{\theta}_1 + \omega_0^2 \delta\theta_1 + 2\gamma\delta\dot{\theta}_1 = \frac{dE}{I} \sin\alpha \cos\omega t$$

Solution

$$\delta\theta_1 = a\cos\omega t + b\sin\omega t$$

where

$$a = -\frac{dE}{2I\omega_0}\sin\alpha \frac{\omega-\omega_0}{(\omega-\omega_0)^2+\gamma^2}\ ,\ b = \frac{dE}{2I\omega_0}\sin\alpha \frac{\gamma}{(\omega-\omega_0)^2+\gamma^2}$$

Absorbed power

$$P = dE\sin\alpha\overline{\cos\omega t\delta\dot{\theta}_1} = \frac{1}{2}dE\sin\alpha\cdot b\omega_0 = \frac{\pi}{4I}d^2E^2\sin^2\alpha\delta(\omega_0-\omega)$$

Second interaction term: Mathieu's equation

$$\delta\ddot{\theta}_2 + \omega_0^2(1 + h\cos\omega t)\delta\theta_2 = 0\ ,\ h = (dE/I\omega_0^2)\cos\alpha$$

Mathieu's equation: periodic and aperiodic solutions; latter increase indefinitely in time; parametric resoances at $\omega = 2\omega_0/n$, $n = 1, 2, 3...$

Thermal fluctuations: wipe out these parametric resonances

All the abve considerations are valid for Classical Dynamics

QM: different! Quantization of the “dipolons”, absorption and emission processes

Note: Static el field E_0 is replaced here by $E_0 = d/2a^3$

Polarization domains; randomness; granular matter; (Maxwell-Wagner-Sillars effect); $\omega_0 = 10MHz$

Discussion. Conclusions

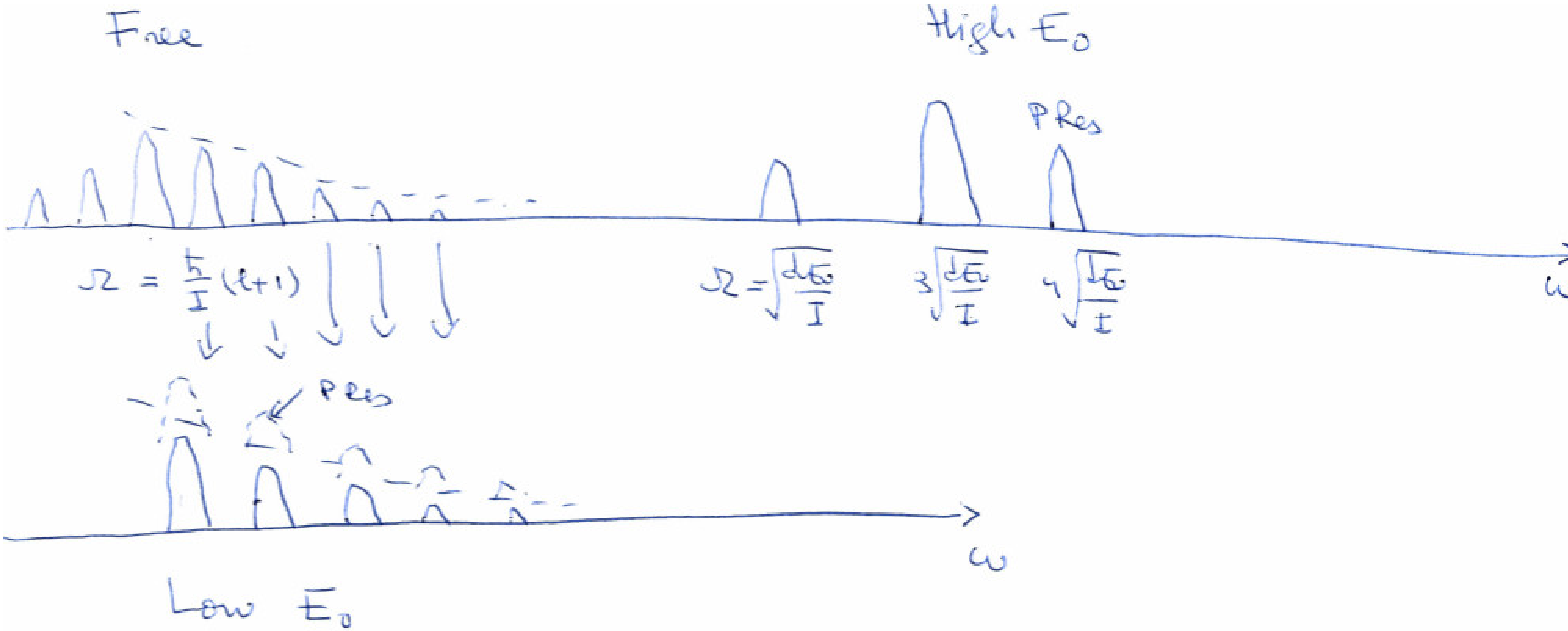
We have shown:

1) Rotations of a molecule (heavy; spherical pendulum) can be approximated by azimuthal rotations and zenithal oscillations

2) Strong static el field (polar matter)$\Longrightarrow$quenched dipoles, parametric resonances

3) Similar in external (weak) static el fields

4) Dipole-dipole interaction $\Longrightarrow$ quenched dipoles, new (polarization) modes, "dipolons" (their excitation$\Longrightarrow$parametric resonance)



Free
High E_0
P Res
$\Omega = \frac{\hbar}{I}(l+1)$
$\Omega = \sqrt{\frac{dE_0}{I}}$
$3\sqrt{\frac{dE_0}{I}}$
$4\sqrt{\frac{dE_0}{I}}$
ω
P Res
ω
Low E_0

5) Extesion to **ferromagnetics**

-Nuclear magn moments $\mu \simeq 10^{-23} erg/Gs$ (5 orders of magnitude less); int energy $\mu^2/a^3 \simeq 10^{-6} K$ practically ineffective

-El magnetic moments $\mu \simeq 10^{-20} erg/Gs$, int energy $\simeq 1K$ (characteristic frequency $\omega_0 \simeq 10^{11} s^{-1}$)

-Note: increase μ by a factor 5, put the nearest neighbours 4, then magnetic dipolar energy increases to $\simeq 100K$, which is near to ferromagnetic transitions temperatures (then, the "magnetic dipolons" become magnons, in ferromagnetic resonances)

Highly-oscillating electric fields

High-power lasers, optical frequency $\omega_h = 2\pi \cdot 10^{15} s^{-1} \gg \omega_{rot,vibr}$

$E_0 \cos \omega_h t$; α highly-oscillating, θ slow ($\alpha \ll \theta$)

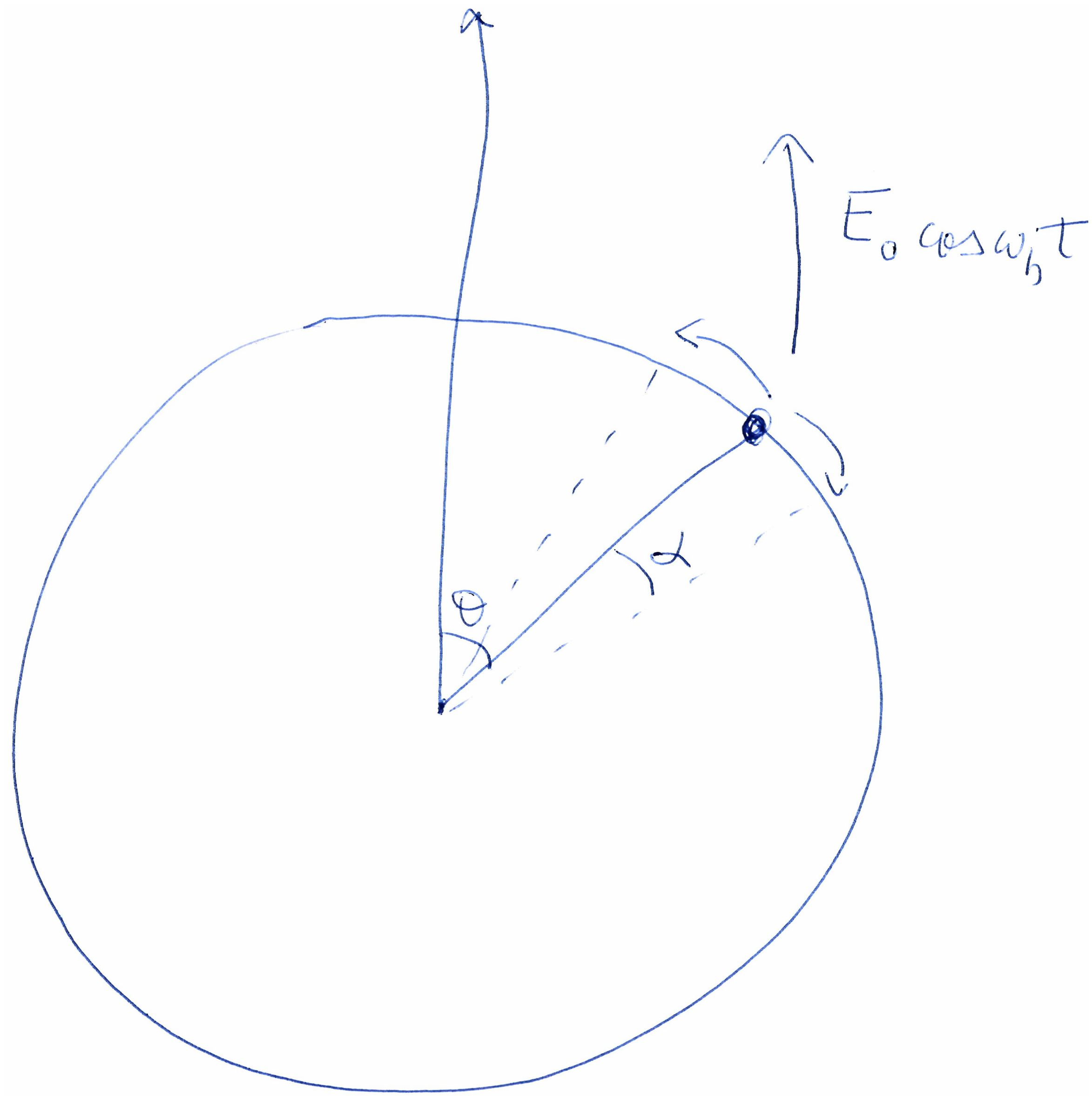
$$I\ddot{\alpha} = -dE_0 \sin(\theta + \alpha) \cos \omega_h t \simeq -dE_0 \sin \theta \cos \omega_h t$$

$$E_{kin} = I\dot{\alpha}^2/2 = (d^2 E_0^2 / 2I\omega_h^2) \sin^2 \theta \cos^2 \omega_h t$$

Average

$$\overline{E}_{kin} = \frac{d^2 E_0^2}{4I\omega_h^2} \sin^2 \theta$$

replaces the interaction energy $-dE_0 \cos \theta$ of the static field in the effective potential energy U_{eff}



$E_0 \cos\omega_L t$
θ
α

$$U_{eff} = \frac{L_z^2}{2I\sin^2\theta} + \frac{d^2E_0^2}{4I\omega_h^2}\sin^2\theta$$

Two minima $\tilde{\theta}_0 = \arcsin\theta_0/R^{1/4}$ and $\tilde{\theta}'_0 = \pi - \tilde{\theta}_0$, $R = dE_0/2I\omega_h^2$ with $\theta_0 = (L_z^2/IdE_0)^{1/4} < R^{1/4}$ for high static fields

Oscillations $\tilde{\omega}_0 = \omega_0\sqrt{3R/4}$, renormalization, $E_0(osc) \to \tilde{E}_0(st) = E_0R$.

Conditions: $\theta_0/R^{1/4} < 1$, $\alpha = (dE_0/I\omega_h^2)\tilde{\theta}_0 \ll \tilde{\theta}_0$ $(R \ll 1)$

$$\frac{\sqrt{2}L_z\omega_h}{d} < E_0 \ll \frac{2I\omega_h^2}{d}$$

With our numerical parameters $I = 10^{-38} g \cdot cm^2$, $T = 300K = 4 \times 10^{-14} erg$, $d = 10^{-18} esu$ and $\omega_h = 2\pi \cdot 10^{15} s^{-1}$:

$$10^8 statvolt/cm < E_0 \ll 10^{10} statvolt/cm$$

$$(R = 10^{-10} E_0 / 2(2\pi)^2 \ll 1)$$

Note: does not affect the translational motion!

Conclude: highly-oscillating electric fields with high intensity like high static electric field, providing renormalization