

Optical Bistability III

Proceedings of the Topical Meeting,
Tucson, Arizona, Dezember 2-4, 1985

Editors: H.M. Gibbs, P. Mandel,
N. Peyghambarian, and S.D. Smith

With 272 Figures

Springer-Verlag
Berlin Heidelberg New York Tokyo

Multiphoton Excitation of Relativistic Cyclotron Resonance and Phase Bistability

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Abstract

Due to relativistic nonlinear effects large amplitude cyclotron motion of a free electron can be excited by two laser beams with frequencies much higher than the cyclotron frequency Ω . The laser frequencies can differ either by Ω (three-photon resonance) or 2Ω (four-photon resonance). The excited cyclotron motion displays a hysteretic resonance based solely on the relativistic mass-effect. In the case of four-photon resonance, the system exhibits novel kind of optical bistability which can be described as phase bistability. The power of laser radiation required to observe these effects is very low and could be obtained using cw regime of e.g. He-Ne (for three-photon resonance) or CO_2 (for four-photon resonance) lasers.

The relativistic effects experienced by a single elementary particle (e.g. electron) oscillating under the action of electromagnetic (EM) field are probably the most fundamental mechanisms of nonlinear interaction of light with matter. We showed earlier^[1] that due to small relativistic changes of mass, a single electron can exhibit pronounced hysteretic cyclotron resonance when the frequency of driving EM field is near the cyclotron frequency. Consistent with this prediction, this effect has recently been observed in experiment^[2] with a single electron trapped for ten months in a Penning trap.

In this paper we show that, due to the combination of the relativistic mass-effect and two other fundamental effects (the Doppler effect and the Lorentz force), still another group of nonlinear optical effects is feasible: large excitation of (microwave) cyclotron motion of an electron by two *optical* waves with their respective frequencies ω_1 and ω_2 ($\omega_1 > \omega_2$) being much higher than the cyclotron frequency Ω ($\omega_1, \omega_2 \gg \Omega$) and differing by either Ω or 2Ω . Hence, these effects may be regarded as three-photon and four-photon interactions, respectively. These multiphoton effects may become a starting point for the entire new field that may be described as a relativistic nonlinear optics of a single electron (or of single particles, in general). The optical frequencies ω_1 and ω_2 can be provided by two modes of the same laser, which allows for easily tunable control over the difference frequency ($\omega_1 - \omega_2$). The power of laser light required to obtain the cyclotron excitation is sufficiently low to allow for the use of laser in a cw or quasi-cw regime. For example, three-photon excitation can be observed using a He-Ne laser with a power as low as a few microwatts.

All of these effects exhibit a relativistic hysteresis, similar in nature to that of a cyclotron resonance which occurs at the main frequency^[1-3], when $\omega = \Omega \approx \Omega_0$. For a four-photon resonance, the new feature is that the excited electron can have *two possible phases* of cyclotron excitation (which differ by π); which phase is excited, depends on the initial conditions. This property may be regarded as a manifestation of a new type of optical bistability which we call *phase bistability* (i.e. that one based on bistability of the phase of oscillation rather than on bistability of its amplitude).

It is known that the conduction electrons in narrow-gap semiconductors demonstrate pseudo-relativistic behavior^[4] governed by the energy-momentum relation (and therefore by the effective mass-energy relation) similar to that for free electrons. Since such an effect may result in hysteretic cyclotron resonance in semiconductors^[5], multi-photon optical excitation of cyclotron resonance in semiconductors, analogous to that of the case of free electrons in vacuum considered here, may also be expected.

Consider a single electron in a homogeneous magnetic field \vec{H}_0 which provides a cyclotron resonance with the initial frequency $\Omega_0 = e H_0/m_0 c$. The electron is illuminated by the optical field which may, in general, consist of any number of plane waves $\vec{E}_j(\omega_j t - \vec{k}_j \vec{r})$ where ω_j and \vec{k}_j are, respectively, the frequencies and wave vectors of the fields. We treat the problem classically; the motion of an electron with an arbitrary momentum $\vec{p} = m_0 \gamma \vec{v}$ under action of any number of plane waves \vec{E}_j is governed by the Lorentz equation (with an additional damping term⁽⁶⁾):

$$\frac{d\vec{p}}{dt} + \Gamma \gamma \Omega_0 \vec{p} = e \sum_j \vec{E}_j + \frac{e}{\gamma m_0 c} \vec{p} \times \sum_j [\vec{k}_j \times \vec{E}_j / k_j] + \frac{e}{\gamma m_0 c} [\vec{p} \times \vec{H}_0] \quad (1)$$

where $\gamma = (1 + p^2/m_0^2 c^2)^{1/2}$ and Γ is a damping parameter due to cyclotron radiation, $\Gamma = 2e^2 \Omega_0 / 3m_0^3 \ll 1$. As distinct from previous work^[1-3] we do not restrict ourselves to the low excitation energies, hence γ can be significantly greater than unity. One may show that in such a case the energy losses described by the second term in the left-hand side of Eq. (1) increase proportionally to $\gamma \vec{p}$ (rather than to just \vec{p} as in the slightly-relativistic case⁽⁶⁾). The second term on the right-hand side in Eq. (1) is the Lorentz force of the incident EM wave which is attributable to the magnetic field of the EM wave, $\vec{H}_j = [\vec{k}_j \times \vec{E}_j] / k_j$. We introduce a dimensionless momentum \vec{p} , fields \vec{E}_j , unit vector of magnetic field \vec{h} , and unit wave vectors \vec{q}_j as follows: $\vec{p} = \vec{p} / m_0 c$; $\vec{E}_j = \vec{E}_j / H_0 = e \vec{E}_j / m_0 c \Omega_0$; $\vec{h} = \vec{H}_0 / H_0$; $\vec{q}_j = \vec{k}_j / k_j$, and assume that the total momentum of the electron can be written in the form

$$\vec{p} = \vec{p}_c(t) + \vec{p}_{nc}^{(1)} + \vec{p}_{nc}^{(2)} + \dots \quad (2)$$

where \vec{p}_c is a "cyclotron" component of momentum describing a pure rotation of the electron around some fixed center ($\vec{r}=0$) with the frequency $\Omega \simeq \Omega_0 / \gamma_c$; \vec{p}_c is orthogonal to \vec{H}_0 . The various orders "noncyclotron" components \vec{p}_{nc} include oscillations with all the other, non-resonant, frequencies and may have any orientation. The cyclotron component \vec{p}_c is determined by the equation:

$$\Omega_0^{-1} (d\vec{p}_c/dt) - \gamma_c^{-1} [\vec{p}_c \times \vec{h}] + \Gamma \gamma_c \vec{p}_c = \vec{F}_c^{(1)}(t) + \vec{F}_c^{(2)}(t) + \dots \quad (3)$$

where $\gamma_c = \sqrt{1 + |\vec{p}_c|^2} = \sqrt{1 + \rho_c^2}$; $\vec{F}_c^{(s)}$ are nonlinear forces of different orders s ; "c" in $\vec{F}_c^{(s)}$ labels those components of these forces that oscillate with the cyclotron frequency Ω and are orthogonal to \vec{H}_0 . In Eqs. (2), (3), $\vec{p}_{nc}^{(s)}$ and $\vec{F}_c^{(s)}$ are defined as:

$$\vec{F}_c^{(1)} = \sum_j \vec{E}_j(\omega_j t - \vec{k}_j \cdot \vec{r}_c(t)) + \gamma_c^{-1} \vec{p}_c \times \sum_j [\vec{q}_j \times \vec{E}_j]; \quad (s=1); \quad (4)$$

$$\Omega_0^{-1} (d\vec{p}_{nc}^{(s)}/dt) - \gamma_c^{-1} [\vec{p}_{nc}^{(s)} \times \vec{h}] + \gamma_c^{-3} (\vec{p}_{nc}^{(s)} \cdot \vec{p}_c) [\vec{p}_c \times \vec{h}] = \vec{F}^{(s)} - \vec{F}_c^{(s)}; \quad (s > 0); \quad (5)$$

$$\vec{F}^{(s)} = \vec{F}_D^{(s)} + \vec{F}_L^{(s)} + \vec{F}_R^{(s)}; \quad (s > 1), \quad (6)$$

where $\vec{r}_c = c \gamma_c^{-1} \int \vec{p}_c dt = -c(\Omega \gamma_c)^{-1} [\vec{p}_c \times \vec{h}]$. Each of the s^{th} order forces $\vec{F}_D^{(s)}$, $\vec{F}_L^{(s)}$, and $\vec{F}_R^{(s)}$ is defined as a sum of all terms of s^{th} order in \vec{E}_j originating, respectively, from the first, second, and third terms on the right-hand side of Eq(1) respectively in which all the lower order terms of \vec{p} in Eq(2) [the highest of which is $\vec{p}_{nc}^{(s-1)}$] are taken into account; note that $\vec{p}_{nc}^{(s)}$ is of s^{th} order in \vec{E}_j . The force $\vec{F}^{(2)}$ is given as follows:

$$\begin{aligned} \vec{F}^{(2)} = & \sum_j (\vec{q}_j \cdot \vec{r}_{nc}^{(1)}) \partial \vec{E}_j / \partial (\vec{q}_j \cdot \vec{r}_{nc}) + \gamma_c^{-1} \vec{p}_{nc}^{(1)} \times \sum_j [\vec{q}_j \times \vec{E}_j] - \\ & \gamma_c^{-3} \{ \vec{p}_{nc}^{(1)} (\vec{p}_n \cdot \vec{p}_{nc}^{(1)}) + (\vec{p}_c / 2) [(\vec{p}_{nc}^{(1)})^2 - 3 \gamma_c^{-2} (\vec{p}_c \cdot \vec{p}_{nc}^{(1)})^2] \} \times \vec{h}; \quad \vec{r}_{nc} = c \gamma_c^{-1} \int \vec{p}_{nc} dt. \end{aligned} \quad (7)$$

In Eq(6) we distinguish *three* main mechanisms of nonlinear interaction, each of which is related to the respective term in Eq(1). The spatial oscillations of the electron make it see the phases $\vec{k}_j \vec{r}$ of the incident fields $\vec{E}_j(\omega_j t - \vec{k}_j \vec{r})$ [the first term in the right-hand side of Eq(1)] rapidly modulated since $\vec{r} = c \gamma^{-1} \int \vec{p} dt$. This modulation is due to the Doppler effect; hence the designation "Doppler" nonlinear mechanism, \vec{F}_D . The Lorentz force [the second term in a right-hand side of Eq(1)] gives rise to components with combination frequencies; hence, the

designation "Lorentz" nonlinear mechanism, \bar{F}_L . Finally, there is a relativistic-mass effect due to $\gamma^{-1} = [1 + (\bar{p})^2]^{-1/2}$ in the last, cyclotron, term on the right-hand side of Eq(1); hence, the designation "relativistic" nonlinear mechanism, \bar{F}_R . Contributions from all these three mechanisms can be of the same order of magnitude. In general, none of them can be neglected, however, for particular propagation and polarization configurations some of them may dominate. It is worth emphasizing, though, that once the cyclotron motion is excited, it is only the relativistic mass-effect [the term γ_c^{-1} on the left-hand side of Eq. (3)] that acts to limit the energy of excitation and to form a hysteretic resonance.

The hierarchical ranking in Eqs(2-6) is tailored in such a way as to emphasize the order of interactions in f_j , not in $(\rho_c f_j)$ (since we assume $f_j \ll 1$, but not necessarily $\rho_c \ll 1$). Due to this fact, the same order term $\bar{F}^{(s)}$ encompasses two (or more) nonlinear interactions that are usually regarded as different orders in conventional nonlinear optics where the polarization of the medium is expressed in powers of the applied field^[7]. Such an "order mixing" is attributable to the finite size r_c of the cyclotron orbit. However, the number of the orders s contributing to any particular nonlinear interaction is always limited and easily found. The force $\bar{F}_c^{(1)}$ is nonzero quantity only for either the main resonance ($\omega = \Omega$) or for the generation of n^{th} order subharmonics ($\omega = n\Omega$); the latter one is a particular case of a multiphoton processes. The subharmonics generation will be discussed elsewhere. The force $F_c^{(2)}$, Eq (7), contributes to three-photon (e.g. $\omega_1 - \omega_2 = \Omega$) and four-photon (e.g. $\omega_1 - \omega_2 = 2\Omega$) resonances both of which are considered below.

Three-photon resonance ($\omega_1 - \omega_2 = \Omega$). Consider two optical waves (with their frequencies $\omega_1 > \omega_2 > 0$) counter-propagating in the direction *orthogonal* to the magnetic field \bar{H}_0 and linearly polarized with their vectors \bar{E}_j *parallel* to \bar{H}_0 . In such a case, in Eq(4), $\bar{F}_c^{(1)} = 0$. In order to find the pumping threshold (which is very low in this case) required to observe relativistic features in the resonance, it is sufficient to consider the weak relativistic case when \bar{v}_c in $\bar{f}_j(\omega_j t - \bar{k}_j \bar{r}_c)$ is small. Assuming, e.g. $\bar{q}_1 = -\bar{q}_2 = \hat{e}_x$, $\bar{h} = \bar{e}_z$, and $\bar{r}_1 = f_1 \hat{e}_x \sin \omega_1 t$, $\bar{r}_2 = f_2 \hat{e}_x \sin \omega_2 t$, we find $\bar{p}_{nc}^{(1)} \approx -\hat{e}_z \Omega_0 (\omega_1^{-1} f_1 \cos \omega_1 t + \omega_2^{-1} f_2 \cos \omega_2 t)$. Substituting this expression in Eq(7) and solving Eq (3) for steady-state cyclotron motion \bar{p}_c , we obtain an equation for the magnitude ρ_c :

$$\rho_c = \mu_{(3)} \{ \Gamma^2 + [(\Omega - \Omega_0)/\Omega_0 + \rho_c^2/2]^2 \}^{-1/2}, \quad (8)$$

where

$$\mu_{(3)} = f_1 f_2 \Omega_0 (\omega_1^{-1} + \omega_2^{-1}) / 4. \quad (9)$$

In this form, Eq(8) is an exact analog of the equation^[1,3] for the *main* resonance when the frequency of driving wave ω is near Ω_0 , the only difference being that instead of a (dimensionless) amplitude μ of the resonant driving wave, we now have a three-photon driving parameter, $\mu_{(3)}$. In order to obtain a hysteresis, which is a distinct signature of a relativistic resonance, one has to have $\mu_{(3)}$ exceed a threshold^[1] $\mu_{cr} \approx 1.75 \Gamma^{3/2}$. Consider the case where $\Omega_0 = 2\pi \times 150$ GHz, i.e. $\lambda_0 = 2$ mm (and therefore the damping parameter $\Gamma \approx 0.6 \times 10^{-11}$), $\lambda_j (= 2\pi/k_j) \approx 0.69 \mu\text{m}$ (He-Ne laser), and $f_1 = f_2$. Making use of Eq(9) we obtain a critical amplitude, $E_{cr} \approx 6$ V/cm, which corresponds to a 48 mW/cm² intensity. If the beam is focused to a spot of $\sim 2 \mu\text{m}$ diameter, this amounts to a total power of only 1.5 μW .

When the driving parameter $\mu_{(3)}$ considerably exceeds the threshold μ_{cr} , the kinetic energy of the cyclotron motion, $\rho_c^2/2$, follows almost exactly the resonant detuning i.e. $\rho_c^2/2 \approx -(\Omega - \Omega_0)/\Omega_0$ (for $\Omega < \Omega_0$), until it reaches its maximum magnitude, $\rho_{c \max}^2/2 = (\mu_{(3)}/\Gamma)^2/2$, which occurs when $(\Omega - \Omega_0)/\Omega_0 = (\mu_{(3)}/\Gamma)/\sqrt{2}$. Immediately after that, if $|\Omega - \Omega_0|$ continues to increase, the electron jumps from the higher excitation branch down to almost zero excitation, curve 1 in Fig. 1a. A perturbation analysis of Eq (3) in the vicinity of steady-states, reveals that the third branch of the steady-state solution, Eq(8), which is located between the higher and lower branches (both of which are stable), is unstable. As the pumping power increases, the electron can be excited to energies above 1 Mev ($\rho_c > 1$).

Four-photon resonance ($\omega_1 - \omega_2 = 2\Omega$). Consider two optical waves counter-propagating along the z axis *parallel* to \bar{H}_0 ; both circularly polarized now in such a way that the polarization of the *higher* frequency (ω_1) wave precesses around \bar{H}_0 in the *same* direction as the electron

motion, whereas the lower frequency (ω_2) wave polarization precesses in the opposite direction. In this case the fields are written as $\vec{f}_j = f_j[\hat{e}_x \sin(\omega_j t + (-1)^j k_j z) - (-1)^j \hat{e}_y \cos(\omega_j t + (-1)^j k_j z)]$; $j=1,2$. One of the interesting features of this case is that it is possible to make an analytic calculation for arbitrary cyclotron energy. Here again, $F_c^{(1)} = 0$. Assume \vec{p}_c in the form $\vec{p}_c = \rho_c[\hat{e}_x \sin(\Omega t + \phi) + \hat{e}_y \cos(\Omega t + \phi)]$, where ρ_c and ϕ are the slowly varying cyclotron momentum amplitude and phase, respectively. Eq (5) yields then: $\vec{p}_{nc}^{(1)} \simeq (\Omega_0/\bar{\omega}) \{[\hat{e}_x \times (\vec{f}_1 - \vec{f}_2)] + \hat{e}_y \rho_c \gamma_c^{-1} [f_1 \sin(\bar{\omega} t - \phi) - f_2 \sin(\bar{\omega} t + \phi)]\}$, where $\bar{\omega} = (\omega_1 + \omega_2)/2$. Substituting this into Eq (7), solving Eq (3) [with a force $\vec{F}^{(2)}$ given by Eq (7)] for a steady-state cyclotron momentum, ρ_c , (or total energy $\gamma_c = \sqrt{1 + \rho_c^2}$), and introducing a four-photon driving parameter $\mu_{(4)}$ as follows:

$$\mu_{(4)} = 2(\Omega_0/\bar{\omega})^2 f_1 f_2 = 2 e^2 E_1 E_2 (c/\bar{\omega})^2 / (m_0 c^2)^2, \quad (10)$$

one arrives at the results given below. Above the threshold, $\mu_{cr} = \Gamma$, the excitation becomes hysteretic and very large; the solution for energy becomes three-valued. One of these solutions is the non-excited state, $\gamma_c = 1$, whereas the two other solutions are given by

$$\gamma_c = \Omega_0/\Omega + (\Omega/\Omega_0)^3 \left\{ -\mu_{(4)}(f_1^2 + f_2^2)/4f_1 f_2 \pm [\mu_{(4)}^2 - \Gamma^2 (\Omega_0/\Omega)^8]^{1/2} \right\}, \quad (11)$$

except for a small gap near Ω_0 where only one of these solutions (the plus sign) exists. For each of the branches [upper, u, with the plus sign in Eq (11), and lower, l, with the minus sign] with $\gamma_c > 1$, there are two solutions for the phase ϕ , given by $\phi_{u,1,2} = -(1/2) \arcsin[(\Gamma/\mu_{(4)})(\Omega_0/\Omega)^4] \pm \pi/2$; and $\phi_{l,1,2} = -\phi_{u,1,2} + \pi/2$. As the frequency Ω decreases, the total energy γ_c increases until it reaches the maximum magnitude $\gamma_{max} = [\mu_{(4)}/\Gamma]^{1/4}$ which occurs when $\Omega = \Omega_{cr} = \Omega_0 \gamma_{max}^{-1}$; immediately after that, the electron jumps to the non-excited state. A perturbation analysis of Eq (3) shows that again the upper branch of the solution, Eq (11), (and phases ϕ_u), is stable, whereas the lower branch (and phases ϕ_l) is unstable. The non-excited solution, $\gamma_c = 1$, is stable everywhere but at a small gap $\pm \Omega_0 \sqrt{\mu_{(4)}^2 - \Gamma^2}$ around $\Omega = \Omega_0 [1 - \mu_{(4)}(f_1^2 + f_2^2)/4f_1 f_2]$. This gap gives rise to the inverse small jump from the non-excited state to the stable excited state, the latter state given by Eq(11) with the plus sign. The existence of two possible stable phases $\phi_{u,1,2}$ allows the electron to retain information concerning the initial conditions of excitation. Such a phase memory suggests an interesting model for a new type of optical bistability which we call phase bistability. We now estimate the critical intensity of laser pumping required to achieve a four-photon cyclotron excitation. Assuming $\lambda_0 = 2$ mm, CO₂ laser ($\lambda_{1,2} \approx 10$ μ m), $E_1 = E_2 = E$, and making use of Eq (10) and critical condition, $\mu_{cr} = \Gamma$, one obtains $E = 0.5 \times 10^4$ V/cm which corresponds to $\sim 0.66 \times 10^5$ W/cm². With a focal spot size of ~ 45 μ m diameter, this corresponds to only 1 W of laser power.

I am indebted to P. L. Kelley for reading this paper. This work was supported by the US Air Force Office of Scientific Research.

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