Isolas in the three-photon optical excitation of a single cyclotron electron

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We show that sufficiently strong laser beams with two frequencies differing by a cyclotron frequency of a free electron can give rise to strong cyclotron excitation with prohibited and allowed cyclotron orbits as well as to an optical Stark shift. This is due to a combination of relativistic effects and results in multiple isolated branches of excited motion (isolas). The formation of the first three-photon isola, when $\lambda_{\text{cyclotron}} \approx 2$ mm and $\lambda_{\text{laser}} \approx 10\ \mu$m, can be observed with the laser power as low as $\cong 10$ mW.

The interaction of microwave and optical radiation with a slightly relativistic single electron can result in strong nonlinear-optical effects.[1–5] These relativistic-based effects constitute the most fundamental mechanism of nonlinear interaction of light with matter[4]; they include hysteresis and bistability in cyclotron resonance of a free electron predicted in Ref. 1 and experimentally observed in Ref. 2, multiphoton resonances,[3] and high-order subharmonic excitation.[5] Most of these effects require a low intensity of driving EM radiation, which is due to small synchrotron-radiation losses of a free electron. By using multiphoton processes,[3,5] the microwave cyclotron resonance of an electron can be excited by *optical* (laser) radiation. In the simplest case of three-photon resonace,[3] two driving laser beams (or modes) whose frequencies differ by a cyclotron frequency can be used for this purpose.

In this Letter we show that when the driving intensity becomes sufficiently high, the three-photon interaction can result in the formation of multiple isolated branches of the electron's kinetic energy as a function of the driving amplitude or frequency detuning (so-called isolas). This suggests that some of the cyclotron orbits are prohibited, which also may be regarded as a forced quantization. We also show that this phenomenon is accompanied by an optical Stark effect, i.e., an intensity-dependent shift of the resonant frequency.

Consider a single electron in a homogeneous magnetic field $\mathbf{H}_0 = \mathbf{h}H_0$, which gives rise to a cyclotron resonance with the initial frequency $\Omega_c = eH_0/m_0c$. The electron is illuminated by *optical* waves whose frequencies are, respectively, ω_1 and ω_2 (with $\omega_1 > \omega_2$). We designate $\omega_1 - \omega_2 = \Omega$ and assume that neither ratio $(\omega_1 + \omega_2)/\Omega$ nor $\omega_{1,2}/\Omega$ is an integer (this excludes higher-order subharmonics[5] and can readily be arranged by the proper frequency tuning). We choose the propagation configuration such that all the traveling waves $\mathbf{E}_j$ propagate in the plane normal to $\mathbf{H}_0$ with their polarizations parallel to $\mathbf{H}_0$ (see the inset in Fig. 1). We describe the motion of the electron by its momentum $\mathbf{p}$ and introduce the dimensionless quantities $\boldsymbol{\rho} = \mathbf{p}/m_0c$, $\mathbf{f}_j = \mathbf{E}_j/H_0 = e\mathbf{E}_j/m_0c\Omega_c$, and $\mathbf{q}_j = \mathbf{k}_j/k_j$, where $k_j = \omega_j/c$. Then a conventional Lorentz equation (which also takes into account an energy loss due to the synchrotron radiation[6]) can be written as[3]

$$\Omega_c^{-1}\dot{\boldsymbol{\rho}} + \Gamma\gamma\boldsymbol{\rho} = \sum_j f_j + \gamma^{-1}\boldsymbol{\rho} \times \left[\sum_j \mathbf{q}_j \times \mathbf{f}_j + \mathbf{h}\right], \quad (1)$$

where $\mathbf{f}_j$ are all the traveling waves participating in the interaction, with their respective arguments being $\omega_j t - \mathbf{k}_j \cdot \mathbf{r}$; $\Gamma = 2e^2\Omega_c/3m_0c^3 <<< 1$ is a synchrotron damping parameter, and $\gamma = (1 + \rho^2)^{1/2}$. Following the procedure of Ref. 3, we expand the momentum $\boldsymbol{\rho}$ as $\boldsymbol{\rho} = \boldsymbol{\rho}_c + \boldsymbol{\rho}_{\text{nc}}^{(1)} + \ldots$, where $\boldsymbol{\rho}_c$ is a pure cyclotron component with the frequency $\Omega \approx \Omega_c/\gamma_c$ and the noncyclotron components of various orders $\boldsymbol{\rho}_{\text{nc}}$. The cyclotron motion is described then by the equation

$$\Omega_c^{-1}\dot{\boldsymbol{\rho}}_c - \gamma_c^{-1}[\boldsymbol{\rho}_c \times \mathbf{h}] + \Gamma\gamma_c\boldsymbol{\rho}_c = \mathbf{F}_c^{(1)} + \mathbf{F}_c^{(2)} + \ldots, \quad (2)$$

where $\mathbf{F}_c^{(s)}$ are cyclotron components of various sth-order forces $\mathbf{F}^{(s)}$, the first two of which are determined by the equations

$$\mathbf{F}^{(1)} = \sum_j \mathbf{f}_j[\omega_j t - \mathbf{k}_j \cdot \mathbf{r}_c(t)] + \gamma_c^{-1}\boldsymbol{\rho}_c \sum_j \mathbf{q}_j \times \mathbf{f}_j, \quad (3)$$

$$\mathbf{F}^{(2)} = \gamma_c^{-1} \sum_j \mathbf{q}_j(\mathbf{f}_j \cdot \boldsymbol{\rho}_{\text{nc}}^{(1)}) - \frac{\boldsymbol{\rho}_c \times \mathbf{h}}{2\gamma_c^3}[\boldsymbol{\rho}_{\text{nc}}^{(1)}]^2, \quad (4)$$

where $\mathbf{r}_c = c\gamma_c^{-1} \int \boldsymbol{\rho}_c dt = -c(\Omega\gamma_c)^{-1}[\boldsymbol{\rho}_c \times \mathbf{h}]$ and

$$\boldsymbol{\rho}_{\text{nc}}^{(1)} = \Omega_c \int \mathbf{F}^{(1)} dt \quad (5)$$

[note that Eqs. (4) and (5) are now valid only for the chosen configuration]. By using Eqs. (3) and (5), it can be directly verified that the first-order noncyclo-

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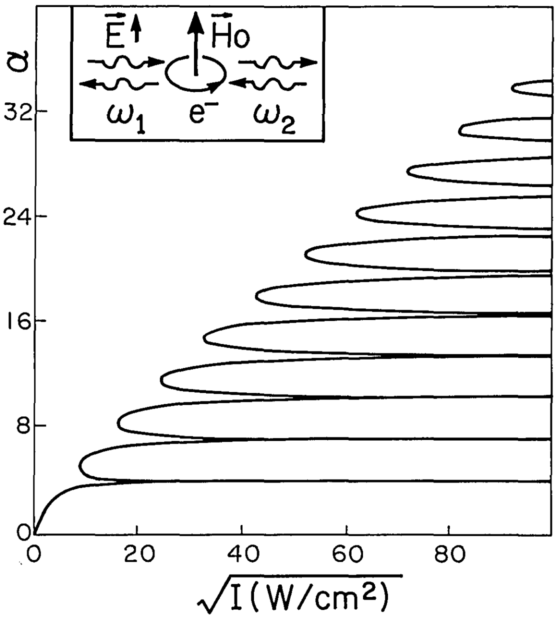

Fig. 1. The maximum and minimum values of excitation parameter α versus the amplitude of driving waves $\sqrt{I}$. The lowest curve corresponds to the maximum of the main hysteresis, the curve above it to the first isola, etc. The area surrounded by each curve corresponds to allowed excitation; the areas between the curves, to prohibited excitation. The inset shows the wave-propagation configuration.

tron momentum $\rho_{nc}^{(1)}$ can be expressed in the amazingly simple form

$$\rho_{nc}^{(1)} = \sum_j (\Omega_c/\omega_j)\mathbf{f}_j(\omega_j t - \mathbf{k}_j \cdot \mathbf{r}_c - \pi/2). \qquad (6)$$

We assume now that $\mathbf{h} = \hat{e}_z$ and that the driving radiation at both frequencies ω_1 and ω_2 forms standing-wave patterns with all the waves propagating along the same axis, say x, i.e., $\mathbf{q}_{n\pm} = \mp\hat{e}_x$; see the inset in Fig. 1. The standing-wave configuration is chosen for the reason that at the equilibrium mode the radiation force acting upon the electron is canceled for both of the waves, and therefore the trapping potential (although weak) can be excluded from the consideration. The electric fields of these standing waves can be expressed as $\mathbf{f}_{\omega_n} = \mathbf{f}_{n+} + \mathbf{f}_{n-}$ ($n = 1, 2$), with $\mathbf{f}_{n\pm} = f_n\hat{e}_z \sin(\omega_n t \pm k_n x + \psi_{n\pm})/2$. Here $\mathbf{f}_{n\pm}$ are counterpropagating traveling waves with the same amplitude $f_n/2$ and frequency ω_n; their phases $\psi_{n\pm}$ in general could be different. The steady state of equilibrium cyclotron motion in the chosen configuration is achieved when the center of the cyclotron orbit coincides with the zero of one of the standing waves and simultaneously with the maximum of the other one. At this point the average radiation forces of all the waves acting on the electron cancel one another. Therefore one of the choices for the phases in the equation for $\mathbf{f}_{n\pm}$ is $\psi_{1+} = 0$, $\psi_{1-} = \pi$, and $\psi_{2\pm} = 0$.

In the three-photon excitation,[3] $\mathbf{F}_c^{(1)} = 0$ in Eq. (2), and, therefore, the first meaningful term contributing to this excitation is $\mathbf{F}_c^{(2)}$. Substituting $\rho_{nc}^{(1)}$ in the form of Eq. (6) into Eq. (4), assuming that ρ_c is in the form $\rho_c = \rho_c[\sin(\Omega t + \phi)\hat{e}_x + \cos(\Omega t + \phi)\hat{e}_y]$, where ρ_c and ϕ are the slowly varying cyclotron momentum amplitude and phase, respectively, separating the cyclotron component $\mathbf{F}_c^{(2)}$ out of $\mathbf{F}^{(2)}$, Eq. (4), and substituting it into Eq. (2), we obtain two scalar equations for the dynamics of ρ_c and ϕ:

$$\gamma_c(\Omega_c^{-1}\dot{\rho}_c + \Gamma\gamma_c\rho_c) = -\mu_{(3)}Q_+ \cos\phi, \qquad (7)$$

$$(\Omega_c^{-1}\dot{\phi} + \Omega/\Omega_c - \gamma_c^{-1})\rho_c\gamma_c = \mu_{(3)}(Q_- - \beta_c^2 Q_+) \times \sin\phi + S\rho_c, \qquad (8)$$

where $Q_\pm$ and S are defined as

$$Q_\pm \equiv J_0(\alpha) \pm J_2(\alpha) + \frac{\Omega}{\omega_1 + \omega_2}[J_0(\beta_c) \pm J_2(\beta_c)], \qquad (9)$$

$$S = \frac{1}{8}\sum_{n=1}^{2} f_n^2 \left\{\left(\frac{\Omega}{\omega_n}\right)^2 [1 + (-1)^n J_0(2\alpha_n)] + 2(-1)^n[J_0(2\alpha_n) + J_2(2\alpha_n)]\right\}. \qquad (10)$$

In Eqs. (7)–(10) $J_\nu(z)$ is the νth-order Bessel function of the first kind, $\beta_c = v/c = \rho_c/\gamma_c$, $\alpha_{1,2} \equiv \omega_{1,2}\rho_c/\Omega_c$, $\alpha = \alpha_1 + \alpha_2$, and $\mu_{(3)}$ is a driving three-photon parameter defined as $\mu_{(3)} = f_1 f_2(\Omega_c/\omega_1 + \Omega_c/\omega_2)/8$. The S term is readily identified as an optical (i.e., intensity-dependent) Stark shift of the cyclotron frequency [see also below, Eq. (12)]. [The signs in front of terms with $(-1)^n$ in Eq. (10) must be changed if $\psi_{1\pm} = 0$, $\psi_{2+} = 0$, and $\psi_{2-} = \pi$.] This shift can become an important factor in future research on the measurement of fundamental constants; it does not, though, cause any significant changes in the structure of the isolas discussed below. The steady state follows from Eqs. (7) and (8) with $d/dt = 0$; in particular, we obtain an equation determining ρ_c as

$$\rho_c^2\left[\frac{\Gamma^2\gamma_c^4}{Q_+^2} + \frac{(\gamma_c\Omega/\Omega_c - 1 - S)^2}{(Q_- - \beta_c^2 Q_+)^2}\right] = \mu_{(3)}^2. \qquad (11)$$

For sufficiently low energy of excitation (i.e., $\alpha_{1,2} \ll 1$, and therefore $\rho_c \approx \beta_c \ll \Omega_c/\omega_{1,2}$ and $r_c \ll \lambda_{1,2}$), Eq. (11) reduces exactly to the equation for the steady-state three-photon excitation [see Eq. (8) of Ref. 3], which results in a hysteretic resonance (curve 1 in Fig. 2) that is typical for other relativistic resonances.[1–5] However, the most interesting new feature following from Eq. (11) is the existence of new, isolated branches of excitation, which we call isolas, that are similar to isolas in other areas of nonlinear physics.[7] In the steady-state solutions, the isolas appear when the driving force $\mu_{(3)}$, and therefore the kinetic energy of the electron, increases. These isolas occur because of a spatially oscillating wave pattern of the driving radiation; they can be expected when $\alpha > 1$.

In order to illustrate the formation of isolas, we consider the case of low-energy excitation $\rho_c^2/2$ (but sufficiently high parameter α). Indeed, since usually $\omega_{1,2} \gg \Omega_c$, the condition $\alpha > 1$ or even $\alpha \gg 1$ can be satisfied even with $\rho_c \approx \beta_c \ll 1$, i.e., still in the low relativistic mode of excitation. In such a case, by simplifying definitions, Eqs. (9) and (10), Eq. (11) can be rewritten as

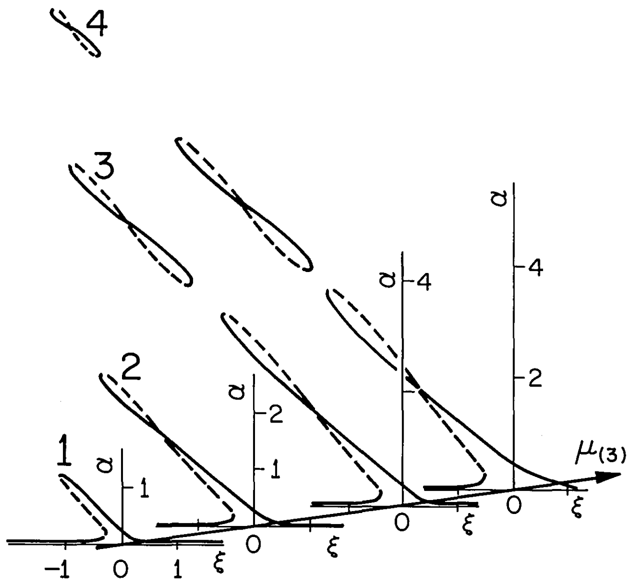

Fig. 2. The excitation parameter α versus frequency-detuning parameter ξ (see the explanation in the text) for the fixed driving parameter $\mu_{(3)}$. Curves: 1, $\mu_{(3)} < \mu_{sc}$ [μ_{sc} is a critical magnitude of $\mu_{(3)}$ for the first self-crossing to occur); 2, $\mu_{(3)} \approx 7\mu_{sc}$ (the first self-crossing appears in main, mother curve); 3, $\mu_{(3)} \approx 2.6\mu_{cr}$ [μ_{cr} is a critical magnitude of $\mu_{(3)}$ for formation of the first isola]; 4, $\mu_{(3)} \approx 4\mu_{cr}$, the formation of the second isola. The solid branches in curves 1–4 correspond to stable states; the dashed ones, to unstable states.

$$\Omega/\Omega_c - 1 = -\rho_c{}^2/2 + S \pm \rho_c{}^{-1}(J_0 - J_2) \times [\mu_{(3)}{}^2 - \Gamma^2\rho_c{}^2\alpha^2/4J_1{}^2]^{1/2}, \qquad (12)$$

where $J_\nu = J_\nu(\alpha)$. It is readily seen that the steady-state excitation is allowed only if

$$2\mu_{(3)}|J_1(\alpha)| \geq \alpha\rho_c\Gamma. \qquad (13)$$

This condition determines the maximum possible momentum of excitation $(\rho_c)_{max}$ for any given driving amplitude $\mu_{(3)}$. Equation (12) shows, on the other hand, that when $\mu_{(3)}$ *exceeds some level*, there are ranges of momentum ρ_c [such that $\rho_c < (\rho_c)_{max}$], in which the steady-state excitation *does not exist*, i.e., some orbits are prohibited (note that $r_c = \rho_c/k_c \approx \beta_c/k_c$, where $k_c = \Omega_c/c$). Equation (13) predicts the sizes of prohibited orbits in the vicinity of zeros of the function $J_1(\alpha)$. For sufficiently large α, the radii of the prohibited cyclotron orbits are $r_{proh} \approx (2l + 1)\bar{\lambda}/8$, where l is an integer and $\bar{\lambda} = 4\pi c/(\omega_1 + \omega_2)$; see Fig. 1.

Prohibited orbits correspond to the destructive interaction of both of the waves with the electron, as opposed to the constructive interaction pertinent to the allowed orbits. This situation gives rise to the isolated branches of solutions {e.g., $\rho_c[\mu_{(3)}]$ and $\rho_c(\Omega)$}, so-called isolas, with different branches separated by $\rho_{proh} = r_{proh}k_c$. As the intensity of the driving waves [or the driving parameter $\mu_{(3)}$] increases, the first isola is formed, then the second, and so on. In Fig. 2 the excitation parameter $\alpha = \rho_c(\omega_1 + \omega_2)/\Omega_c$ is depicted versus frequency-detuning parameter $\xi = \text{sign}(\Delta)\sqrt{|2\Delta|}(\omega_1 + \omega_2)/\Omega_c$ (where $\Delta \equiv \Omega/\Omega_c - 1$ and "sign" is the sign function) for different fixed magnitudes of $\mu_{(3)}$.

The results of the stability analysis of Eqs. (7) and (8) are shown in Fig. 2, in which the solid lines of the steady states correspond to stable branches and the dashed lines to unstable branches. The critical magnitude of driving parameter $\mu_{(3)}$ required for observation of the excitation of the nth isola is determined by Eq. (13) (with the equality sign), in which for α one has to substitute the nth positive root of the equation, $J_0(\alpha) + 3J_2(\alpha) = 0$. The first isola corresponds at $\alpha \approx 5$ and requires that $\mu_{(3)} = \mu_{cr} \approx 38[\Omega_c/(\omega_1 + \omega_2)]\Gamma$. Assuming that $\lambda_c \approx 2$ mm, $\lambda_{1,2} \approx 10$ μm (CO_2 laser), and that the driving intensity is equal at both of the laser frequencies ($f_1 = f_2$), we estimate the critical laser intensity as ≈ 77 W/cm^2. With an illuminated spot $\lambda_c \times \lambda_{laser} \approx 10^{-4}$ cm^2, this translates into the total power $\sim 10^{-2}$ W.

Figure 2 shows that a curious feature of all these regimes is the self-crossing of steady-state solutions that occurs both in the isolas (curves 3 and 4 in Fig. 2) and in the main, mother curve (curve 2 in Fig. 2). One should note, though, that the point of self-crossing actually corresponds to two *different* states of the system with *different* phases ϕ, which are readily found from Eqs. (7) and (8) with $d/dt = 0$. Equation (12) shows that the self-crossings occur at ρ_c, determined by the equation $J_0(\alpha) = J_2(\alpha)$, i.e., at $r_{sc} \approx \bar{\lambda}l/4$, where l is an integer (for sufficiently large l). The critical magnitude of $\mu_{(3)}$ for the first self-crossing to occur is $\mu_{sc} \approx 3[\Omega_c/(\omega_1 + \omega_2)]\Gamma \approx \mu_{cr}/13$, which, in the above-mentioned case, corresponds to the laser intensity ≈ 6 W/cm^2. Experimentally, both hysteresis and isolas may be observed by measuring either the mass of electrons[2] or radiation at the cyclotron frequency Ω or the four-wave mixing frequencies $\omega_3 = 2\omega_1 - \omega_2$ and $\omega_4 = 2\omega_2 - \omega_1$.[8]

In conclusion, we have demonstrated that the three-photon interaction of a biharmonic laser with a single electron may result in strong cyclotron excitation, characterized, in addition to relativistic hystereses, by multiple isolas. The critical power of laser radiation required for observation of the first isola is very low and can be obtained by using the cw regime of any conventional laser.

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