Hysteretic three-photon cyclotron resonance in semiconductors

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We predict a hysteretic three-photon cyclotron resonance in narrow-gap semiconductors driven by two laser beams with their frequency difference near the cyclotron frequency. This effect is based on the Doppler and Lorentz nonlinear mechanisms and the nonparabolicity of the conduction band. A CO_2 laser intensity of 10^5–10^6 W/cm^2 at 10.6 and 9.4 μm is required for observation of the effect at 83.03 μm in InSb, GaAs, or HgTe.

In previous work[1] by one of us it was shown that because of relativistic effects a large cyclotron motion of a free electron in vacuum can be excited by two laser beams with their frequencies much higher than a cyclotron frequency Ω. The laser frequencies must differ by either Ω or 2Ω, which correspond to three-photon or four-photon resonance, respectively. Although these multiphoton resonances can be caused by three different mechanisms (which were identified[1] as the Doppler, Lorentz, and relativistic mechanisms), the excited motion always displays a hysteretic resonance based solely on the relativistic mass effect. The hysteretic resonance of a slightly relativistic electron at the main frequency was predicted by one of us[2] and subsequently observed experimentally.[3] It was also shown[2,4] that the hysteretic resonance of a similar nature at the main frequency may occur in narrow-gap semiconductors owing to the pseudorelativistic properties of their conduction electrons.

In this Letter we consider the feasibility of a hysteretic three-photon resonance in narrow-gap semiconductors analogous to that of a free electron in vacuum.[1] The hysteresis is feasible because of the nonparabolicity of the semiconductor conduction band, which causes a pseudorelativistic dependence[5,6] of the effective mass of the conduction electrons on their momentum or energy. The generation of a difference frequency $\Omega = \omega_1 - \omega_2$ by using the nonparabolicity of a semiconductor by laser beams with frequencies ω_1 and ω_2 has already been demonstrated in the earlier work using either spin resonance[7] or cyclotron resonance.[7,8] However, the hysteretic resonance was not observed (or looked for), which might be attributed to a laser intensity apparently insufficient for such an effect. In this Letter we show that this effect is theoretically feasible in narrow-gap semiconductors such as InSb, GaAs, and HgTe, driven by two modes of a CO_2 laser at 10.6 and 9.4 μm such that the difference frequency corresponds to $\lambda \sim 83$ μm, with the driving intensity being 10^5–10^6 W/cm^2.

Following the Kane two-band model[5] with isotropic bands, the energy of the conduction electrons in narrow-gap semiconductors can be expressed as $W(p) = (m^*{}_0{}^2 v_0{}^4 + p^2 v_0{}^2)^{1/2}$, where $\mathbf{p}$ is the momentum of the conduction electron, $m^*{}_0$ is its effective mass at the bottom of the conduction band, $v_0 = (W_G/2m^*{}_0)^{1/2}$ is some characteristic speed, and W_G is the band gap (the energy W is measured with respect to the middle of the gap). Since the velocity $\mathbf{v}$ of the conduction electron is given by[5] $\mathbf{v}(\mathbf{p}) = \partial W(p)/\partial \mathbf{p}$, this yields

$$\mathbf{v} = \mathbf{p}/m^*{}_0(1 + p^2/p_0{}^2)^{1/2}, \tag{1}$$

where $p_0 = m^*{}_0 v_0 = (W_G m^*{}_0/2)^{1/2}$ is some characteristic momentum. One can see [Eq. (1)] that relations among W, v, and p are completely relativisticlike, with v_0 posing as an effective speed of light and $W_G/2$ as an effective rest energy of the electron. The motion of an electron in the semiconductor layer under the action of plane EM waves is governed by the relaxation-modified Lorentz equation

$$\frac{d\mathbf{p}}{dt} + \frac{1}{\tau}\mathbf{p} = e\sum_j \mathbf{E}_j + \frac{e}{c}\mathbf{v} \times \left(\mathbf{H}_0 + \sum_j \mathbf{H}_j\right), \tag{2}$$

where τ is the relaxation time of the momentum, which depends on the scattering of electrons, e is the electron charge, $\mathbf{H}_0$ is a homogeneous dc field, and $\mathbf{E}_j$ and $\mathbf{H}_j$ are, respectively, the electric and magnetic fields of the EM waves with all frequencies ω_j. For plane waves $\mathbf{H}_j = \sqrt{\epsilon_j}(\mathbf{k}_j/k_j \times \mathbf{E}_j)$, where $\epsilon_j = \epsilon(\omega_j)$ is the dielectric constant at the frequency ω_j.

Consider now three-photon excitation ($\Omega = \omega_1 - \omega_2$) for the configuration when two plane waves with their respective frequencies ω_1 and ω_2 counterpropagate normally to $\mathbf{H}_0$ (e.g., along the axis x, i.e., $\mathbf{q}_1 = -\mathbf{q}_2 = \mathbf{e}_x$, where $\mathbf{q}_{1,2} = \mathbf{k}_{1,2}/k_{1,2}$ and $k_{1,2} = \omega_{1,2}/c$) with their polarizations parallel to $\mathbf{H}_0$, and assume that $\mathbf{H}_0 = H_0\mathbf{e}_z$ (see the inset of Fig. 1). We shall follow the approach in which the momentum $\mathbf{p}$ is assumed to be approximately a sum of a pure cyclotron component $\mathbf{p}_c$ with a frequency of rotation $\Omega = \Omega_0/\gamma_c$ [where $\gamma_c = (1 + p_c{}^2/p_0{}^2)^{1/2}$ and $\Omega_0 = eH_0/m^*{}_0 c$] and the first-order noncyclotron momentum $\mathbf{p}_{nc}{}^{(1)}$, which gives rise to the second-order nonlinear force $\mathbf{F}^{(2)}$. Then, from Eq. (2), the motion $\mathbf{p}_c$ is governed by the equation

$$\Omega_0{}^{-1}(d\mathbf{p}_c/dt) - \gamma_c{}^{-1}[\mathbf{p}_c \times \mathbf{e}_z] + \Omega_0{}^{-1}\tau^{-1}\mathbf{p}_c \cong \mathbf{F}_c{}^{(2)}(t), \tag{3}$$

where $\mathbf{F}_c{}^{(2)}$ is the cyclotron component of $\mathbf{F}^{(2)}$. To find a threshold of hysteretic resonance with comparatively low energy and momentum, i.e., $r_c k_j \ll 1$, we assume that $\gamma_c \approx 1$ and obtain $\mathbf{F}^{(2)}$ as

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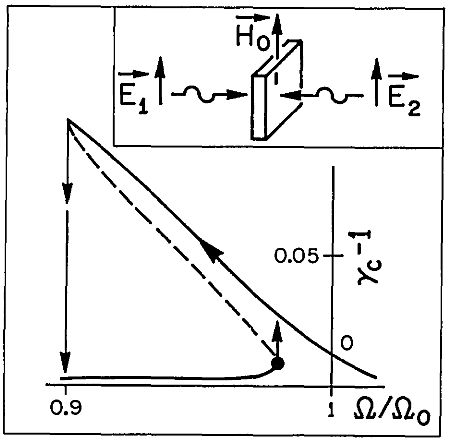

Fig. 1. Plot of kinetic energy $\gamma_c - 1$ of the cyclotron electron versus the effective driving frequency $\Omega = \omega_1 - \omega_2$ in the case $\mu_{(3)}/\Gamma = 0.45$. The solid lines correspond to stable states, and the dashed line to unstable states. The insert depicts polarizations of laser fields $\mathbf{E}_1$ and $\mathbf{E}_2$ with respect to the magnetic dc field $\mathbf{H}_0$ and the thin semiconductor film.

$$\mathbf{F}^{(2)} = -\frac{e}{m^*_0 c\Omega_0 p_0}\sum_j \sqrt{\epsilon_j} \times \left\{[\mathbf{q}_j \cdot \int \mathbf{p}_{\mathrm{nc}}^{(1)} \mathrm{d}t] \frac{\partial \mathbf{E}_j}{\partial t} - \mathbf{q}_j[\mathbf{E}_j \cdot \mathbf{p}_{\mathrm{nc}}^{(1)}]\right\} . \quad (4)$$

In this approximation, only the Doppler and Lorentz forces[1] are responsible for the nonlinear excitation. However, once the cyclotron motion is excited, it is only the mass effect[1] that acts to limit the excitation and to form a hysteresis. Assuming that the driving waves are in the form $\mathbf{E}_{1,2} = \mathbf{e}_z E_{1,2} \sin(\omega_{1,2}t \mp k_{1,2}x_c)$, $\mathbf{p}_{\mathrm{nc}}^{(1)}$ in Eq. (4) can be expressed as

$$\mathbf{p}_{\mathrm{nc}}^{(1)} = -[(eE_1/\omega_1)\cos(\omega_1 t - k_1 x_c) + (eE_2/\omega_2)\cos(\omega_2 t + k_2 x_c)]\mathbf{e}_z. \quad (5)$$

Inserting this into Eq. (4) and assuming[9] that $k_j x_c \ll 1$, we obtain $\mathbf{F}_c^{(2)} = e^2 E_1 E_2(\sqrt{\epsilon_1}/\omega_1 + \sqrt{\epsilon_2}/\omega_2)\sin \Omega t \mathbf{e}_x / 2m^*_0\Omega_0 c p_0$. Assuming now that $\mathbf{p}_c = p_c[\mathbf{e}_x \sin(\Omega t + \phi) + \mathbf{e}_y \cos(\Omega t + \phi)]$ and inserting $\mathbf{F}_c^{(2)}$ into expression (3), we obtain the relationship determining all possible steady-state magnitudes of momentum p_c:

$$p_c/p_0 = \mu_{(3)}[\tau^{-2}\Omega_0^{-2} + (\Omega/\Omega_0 - 1 + p_c^2/2p_0^2)^2]^{-1/2}, \quad (6)$$

where $\mu_{(3)}$ is a dimensionless three-photon driving parameter,

$$\mu_{(3)} = \frac{1}{4}\frac{c}{v_0}(m_0/m^*_0)^2 \frac{e^2 E_1 E_2}{m_0^2 c^2 \Omega_0}(\sqrt{\epsilon_1}/\omega_1 + \sqrt{\epsilon_2}/\omega_2), \quad (7)$$

which differs from the equation in Ref. 1 by the factors v_0/c and $\sqrt{\epsilon_j}$. In narrow-band semiconductors, $m^*_0 \ll m_0$ and $v_0 \ll c$, which implies, first, that the cyclotron resonance can be observed for much higher frequencies (compared to free electrons) for the same magnetic field; for the fields currently available, it could be done in the infrared range. Second, the nonlinearity that is due to the pseudorelativistic mass effect is scaled by c/v_0 and becomes much larger than the relativistic mass effect of free electrons. Under appropriate conditions, Eq. (6) has three solutions. A small-perturbation analysis reveals that these solutions with the higher and lower magnitudes of p_c are stable, whereas the intermediate solution (see Fig. 1) is unstable, which results in a bistable (and hysteretic) behavior of steady-state excitation.[10] It follows from Eq. (6) that, in order to obtain a hysteresis, one has to have $\mu_{(3)}$ exceed a threshold

$$\mu_{\mathrm{cr}} \approx 1.75(\tau\Omega_0)^{-3/2}. \quad (8)$$

To estimate the critical laser intensity determined by expression (8), we consider three different semiconductors (InSb, GaAs, and HgTe) pumped by two lines of a CO_2 laser at wavelengths $\lambda_1 = 10.6$ and $\lambda_2 = 9.4$ μm with each having the same intensity (i.e., $E_1 = E_2$), such that the difference frequency is located in a far-infrared range, $\lambda_0 = (\lambda_2^{-1} - \lambda_1^{-1})^{-1} = 83.03$ μm. The effective mass ratio m^*_0/m_0 is 0.015,[11] 0.016,[12] and 0.029 (Ref. 13) for InSb, GaAs, and HgTe, respectively; therefore the magnetic field H_0 resulting in the cyclotron wavelength $\lambda_0 = 83$ μm is 1.95, 8.51, and 3.74 T, respectively. The temperatures required for these semiconductors are 4.2, 77, and 10 K, whereas their energy gaps W_G are 0.237,[11] 1.54,[12] and 0.283 (Ref. 11) eV, respectively. The dielectric constants[12] are 15.7, 10.9, and 19, respectively (we assume that the dielectric constant is the same at ω_1 and ω_2 since these frequencies are close). For these three cases, we assume that the relaxation time τ is roughly the same, $\tau \approx 10^{-11}$ sec, which was directly measured for InSb (Ref. 11) and can be obtained from the data on the Hall mobility μ for GaAs (Ref. 11) and HgTe (Ref. 13) as $\tau = m^*_0\mu/e$. When evaluating the critical intensity of the laser field through Eq. (7) and expression (8), we notice that the microscopic fields E_j in Eq. (2) are related to the macroscopic laser field E_L by the Clausius–Mossotti formula $E_j = E_L(\epsilon_j + 2)/3$. The resulting critical laser intensity required to obtain hysteresis is 4.3×10^4, 1.64×10^6, and 1.54×10^5 W/cm^2 for InSb, GaAs, and HgTe, respectively.

Other nonlinear effects (first, the *interband* multiphoton processes in the case of InSb) may provide a strong competition to the nonlinear *intraband* processes under consideration. The analysis of the experiment and data[8] clearly shows, however, that in the required range of laser intensities, the interband two-photon and three-photon absorption could be neglected. Indeed, the critical intensity for InSb is 4.3×10^4 W/cm^2; this is less than the 10^5 W/cm^2 used in the experiment,[8] which also used nonparabolicity of the conduction band.[14] Therefore in the range of intensities $\lesssim 10^5$ W/cm^2 the intraband nonparabolic effects in InSb should dominate the interband multiphoton effects, which makes it feasible to observe pseudorelativistic hysteresis for appropriately chosen samples of InSb.

Another material, HgTe, at first glance might seem inappropriate to use since it is known as a zero-gap material with a negative energy gap.[15] The nonparabolicity of the conduction band in HgTe, however, can still be approximated by Kane's model, whereby W_G in Eq. (1) is to be interpreted[15] now as the minimum energy difference between valence bands Γ_6 and Γ_8. Indeed, it was shown empirically[15] that the effective mass can be expressed then as $m^* \approx m^*_0(1 + 2E/W_G)$, where E is the kinetic energy of an electron, which comes exactly to Eq. (1) (for $E/W_G \ll 1$). The two-photon transition between valence bands may result in the increased absorption of the pumping laser radiation, which should apparently be taken into consideration when one is designing an experiment with HgTe.

The attractive feature of GaAs is the relatively high temperature (~77 K) sufficient for the experiment. In addition, GaAs does not suffer from the interband multiphoton processes since the energy gap is quite large compared with 0.1 eV, corresponding to the CO_2 laser energy. The nonparabolicity is consequently small, and therefore the laser threshold intensity is high compared with that of other materials. However, the considerable excitation-related change of the effective mass of the conduction electron has been observed in the most recent experiment.[16] A feasible perturbing mechanism in GaAs, which is a polar crystal, is the excitation of optical photons by laser radiation. The recent experiment[17] showed, however, that the optical phonons in GaAs cannot be excited even at $\lambda \sim 1$ μm, i.e., they do not exist for longer wavelengths, in particular, for the CO_2 laser.

Experimentally, the hysteresis of cyclotron resonance may be observed both by measuring the current or radiation at the cyclotron frequency Ω and the radiation at the optical frequencies $\omega_3 = 2\omega_1 - \omega_2$ and $\omega_4 = 2\omega_2 - \omega_1$ attributable to the four-wave mixing that is due to pseudorelativistic nonlinearity. Indeed, Eq. (5) shows that for the low-energy excitation [$p_c \ll m^*_0 c\Omega_0/(\omega_j\sqrt{\epsilon_j})$], the noncyclotron momentum $\mathbf{p}_{nc}^{(1)}$ at frequencies ω_3 and ω_4 is $\mathbf{p}_{nc}^{(1)}(\omega_{3,4}) \approx \pm\hat{e}_z(eE_{1,2}\sqrt{\epsilon_{1,2}}/2m^*_0\Omega_0 c)p_c \sin(\omega_{3,4}t \pm \phi)$. This implies that the electron current $\sim\mathbf{p}_{nc}^{(1)}(\omega_{3,4})$ oscillating at frequencies $\omega_{3,4}$ along magnetic field $\mathbf{H}_0$ is proportional to p_c [Eq. (6)] and therefore must directly reflect the three-photon hysteresis. This provides a simple method for the observation of three-photon hysteresis. Polarizations of counterpropagating beams of a CO_2 laser with wavelengths 10.6 and 9.4 μm are chosen parallel to the magnetic field $\mathbf{H}_0$, which in turn is directed in the plane of the thin semiconductor sample, with the laser beams incident normally upon the sample [which is a special case of the Voigt configuration (see Ref. 8 and the inset of Fig. 1)]. Equation (5) predicts that radiation at both frequencies ω_3 and ω_4 will be polarized parallel to $\mathbf{H}_0$ and that the ratio of the intensity of radiation at those frequencies will be $E_1^2\omega_3^2/E_2^2\omega_4^2$; the intensity of this radiation is to undergo hysteresis directly related to p_c.

In conclusion, we have demonstrated the feasibility of hysteretic (bistable) three-photon resonance in narrow-gap semiconductors using two laser beams with their difference frequency near the cyclotron frequency. By using two lines of a CO_2 laser, a large hysteretic excitation may be observed in a far-infrared range. This effect may be of significant interest both for the study of highly excited Landau levels and for such applications as resonatorless optical bistability[18] and far-infrared sources of coherent radiation.

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