NONLINEAR MAGNETO-OPTICS OF LANDAU ELECTRONS

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(Received 12 September 1966)

In the parabolic band model, the intraband nonlinear resonance and tunneling of Landau electrons in the presence of a strong electromagnetic field can be calculated exactly to all orders in the electric field strength.

The recent development of lasers as the source of intense, coherent, and monochromatic electromagnetic radiation presents many interesting possibilities for studying nonlinear effects in atoms and solids. Theoretical and experimental studies on nonlinear processes including multiphoton resonances, tunneling, and higher harmonics generation in the absence of an applied magnetic field have been reported by Braunstein,[1] Inoue and Toyozawa,[2] Kelly,[3] Keldysh,[4] and others.[5] In this Letter, we wish to report a theoretical study on the nonlinear response of Landau electrons in a strong electromagnetic field. Because the output frequency of a laser is in general fixed, we find it useful to investigate the nonlinear effects in the presence of a static applied magnetic field. In this case, the magnetic field strength (the cyclotron frequency) can be varied to probe the detailed structure of nonlinear effects even though the laser frequency is fixed. Very recently, Button et al.[6] have observed multiphoton absorption peaks in InSb and PbTl by sweeping the applied magnetic field. These peaks have been attributed to interband transitions between the Landau states of valence and conduction bands.

Because an exact theoretical approach to the problem of nonlinear magneto-optics has not been offered, one is usually forced to adopt a suitable perturbation or iterative technique for analysis of experimental data. There exists, however, a very special case, namely, the case of intraband transitions in the parabolic band model, for which we have been able to obtain exact theoretical results to all orders in the electric field strength in analytic form, whereas we have not been able to do the same for nonparabolic bands and interband transitions. These results are discussed below.

In the case of intraband transitions with crossed electric and magnetic fields, the problem is to find the exact time-dependent wave function for the Hamiltonian,

$$H(t)=p_x^{\,2}/2m+\tfrac{1}{2}m\omega_c^{\,2}(x-X_0)^2-eE_0x\sin\omega t \qquad (t\geq 0), \tag{1}$$

where $X_0=-p_y/m\omega_c$ is the Landau orbit center in the absence of the electric field, $\omega_c=eH/mc$ is the cyclotron frequency, $E(t)=E_0\sin\omega t$ is the (nearly) homogeneous[7] oscillating electric field of frequency ω. [Here, we use the Landau gauge $\vec{A}=(0,-xH,0)$ for static magnetic field H parallel to the z axis, and, for convenience, omit the state labels (p_y,p_z) which are unaffected in the magneto-optical transitions of interest.] Considering this as a time-development problem in which the electromagnetic field is switched on at $t=0$, we solve for the analytic form of the time-ordered wave function,

$$\psi_n(x,t)\rangle=U(t,0)\psi_n(x,0)\rangle$$
$$=(\exp[-i/\hbar\int_0^t H(\tau)d\tau]\psi_n(x,0)\rangle)_+, \tag{2}$$

which describes the behavior at $t>0$ of an electron which was initially at a given Landau state $n(=0,1,2,\cdots)$. Here, $\psi_n(x,0)=\varphi_n(x-X_0)$ is the Hermite function of order n with orbit center at X_0. In Eq. (2), the time-development operator $U(t,0)$ has its operator properties with respect to x and p_x only. Therefore, in order to calculate the wave function in analytic form to all orders in E_0, we proceed by searching for another more convenient unitary operator $S=\exp[iF(x,p_x,t)]$, in which the time integral (and hence the time ordering) has been completed to all orders in E_0 while it satisfies all properties of the operator $U(t,0)$ such that (a) $S^{-1}(x(0),p_x(0))S\equiv U^{-1}(x(0),p_x(0))U\equiv(x(t),$

$p_x(t)\rangle$, and (b) $\psi_n(x,t)\rangle = S\psi_n(x,0)\rangle$. By solving (a) we obtain[8]

$$\exp[iF(x,p_x,t)] = \exp[i(x-X_0)\alpha(t)/\hbar \exp[-ip_x\beta(t)/\hbar] \times \exp\{-i[p_x^2/2m + \tfrac{1}{2}m\omega_c^2(x-X_0)^2]t/\hbar\}, \quad (3)$$

aside from the phase factor which is independent of x, p_x, and n and, therefore, is omitted, and where $\alpha(t)$ and $\beta(t)$, when normalized to be dimensionless, are given by

$$\alpha(t) = \frac{eE_0}{2}\left(\frac{1}{m\hbar\omega_c}\right)^{1/2} \times \left\{\frac{1-\cos(\omega_c+\omega)t}{\omega_c+\omega} + \frac{1-\cos(\omega_c-\omega)t}{\omega_c-\omega}\right\},$$

$$\beta(t) = \frac{eE_0}{2}\left(\frac{1}{m\hbar\omega_c}\right)^{1/2}\left\{\frac{\sin(\omega_c+\omega)t}{\omega_c+\omega} + \frac{\sin(\omega_c-\omega)t}{\omega_c-\omega}\right\}. \quad (4)$$

In (3), the exponential factor containing $\alpha(t)$ has the effect of mixing different Landau states whose orbit centers oscillate with the sum and difference frequencies $(\omega_c \pm \omega)$ in the manner of (4). These oscillatory modes result from beating of the cyclotron motion and frequency ω of the driving field.

For a given initial state n, the resonance and tunneling amplitudes can be readily obtained by calculating the composition, $C_{ln}(t) = \langle\varphi_l(x-X_0), \psi_n(x,t)\rangle$ for various Landau states, (l, p_y, p_z). For convenience, let us consider the simple case of absolute zero temperature at $t=0$ and the Fermi energy $\zeta < \frac{1}{2}h\omega_c$ such that initially, all electrons were at the Landau ground state $n=0$. In order to investigate the resonance properties, we evaluate the rate of transition probability W_l in the long-time limit $t(\gg\omega^{-1}, \omega_c^{-1}) \to \infty$ and obtain

$$W_l = \lim_{t\to\infty}\frac{\partial}{\partial t}|C_{l,0}(t)|^2 \quad (l\neq 0),$$
$$= 2\pi\delta(\omega_c \pm \omega)\left(\frac{eE_0\lambda}{2\hbar}\right)^2 \frac{1}{(2l-2)!}\left(\frac{eE\pm\lambda}{2\hbar}\right)^{2l-2} \times \left\{1-\frac{1}{2l}\left(\frac{e\lambda E\pm}{2\hbar}\right)^2\right\}\exp\left\{-\frac{1}{2}\left(\frac{e\lambda E\pm}{2\hbar}\right)^2\right\}, \quad (7)$$

where $\lambda = (\hbar/m\omega_c)^{1/2}$ is the orbit radius of Landau ground state, and $E_\pm$ denote the Fourier components of the oscillating electric field for $\omega = \mp\omega_c$. Observe that, while this contains terms to all orders in E, only the one-photon (cyclotron) resonances are allowed and multiphoton resonances do not appear. This implies that the resonance transitions to higher Landau states take place by ladderlike steps of one-photon resonances only. The multiphoton resonances do not appear in the present result because, in the pure harmonic oscillator system (i.e., cyclotron motion in the parabolic band), there exists no nonlinear polarization source.[9] Note that, in Eq. (7), a change in sign occurs when $l(\neq 0)$ is equal to $l_0 = \frac{1}{2}(e\lambda E\pm/2\hbar)^2$. This implies that, when the electric field is strong enough to make $l_0 > 1$, electrons at the states $l < l_0$ are steadily pumped up into the higher states $l > l_0$ by action of the strong electromagnetic radiation. This process will continue until the rate (7) is counterbalanced, for example, by the rate of downward transitions due to collisions with phonons.

In the case of off-resonance conditions, the tunneling transitions to higher Landau states are possible when the electric field strength is sufficiently large. This, however, is a transient phenomenon and the rate $\partial|C_{l,0}(t)|^2/\partial t$ of pure tunneling vanishes at large t. In the limit $t\to\infty$, nevertheless, the tunneling gives rise to steady-state values of population which are given by

$$\lim_{t\to\infty}|C_{l,0}(t)|^2 = \frac{1}{2^l l!}\left[\frac{eE_0\lambda\omega}{\hbar(\omega_c^2-\omega^2)}\right]^{2l}\exp\left\{-\frac{1}{2}\left[\frac{eE_0\lambda\omega}{\hbar(\omega_c^2-\omega^2)}\right]^2\right\}, \quad (8)$$

when $\omega_c^2 \neq \omega^2$. The tunneling mechanism responsible for the above result is illustrated schematically in Fig. 1 and may be explained as

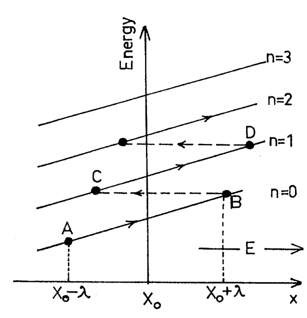

FIG. 1. Tunneling of electrons between two Landau levels.

follows: When $\omega \ll \omega_c$, the electrons move in a nearly static electric field and the energy difference between the sites A and B in Fig. 1 is approximately $2eE_0\lambda$. If E_0 is large enough to satisfy the condition $(2eE_0\lambda) > \hbar\omega_c$ (which can be easily realized in a microwave cavity or by infrared laser), it is possible for electrons at the lth Landau level to tunnel into the $(l+1)$th level. For this condition, the average Landau quantum number $\langle l \rangle$ is calculated to be $\frac{1}{2}\{eE_0\lambda/\hbar\omega_c[1-(\omega/\omega_c)^2]\}^2$, which can be a very large number for strong E and/or ω near the resonance. In the above calculations, we have neglected any mechanism of relaxation assuming that, because of strong E, the laser-induced transition rates are much greater than the inverse relaxation time. Once the laser is shut off, however, the excited electrons may cascade down to the lower states via scattering (e.g., phonons) as well as by spontaneous emission of photons.

Finally, a few words about intraband transitions in nonparabolic bands. Perturbation theoretical considerations show that, unlike in parabolic band model, multiphoton resonances between Landau subbands are possible. When the energy minimum of the conduction band is situated at the Brillouin zone point which has the inversion symmetry, only the odd multiphoton processes are allowed. Otherwise, all multiphoton processes may be allowed. This can be deduced from the effective mass representation of the Hamiltonian. Further details on the present theoretical study will be published separately as a paper.

It is a pleasure to thank Dr. P. N. Argyres for very helpful discussions.

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*On leave of absence from the University of Tokyo, Tokyo, Japan.

†Supported by the U. S. Air Force Office of Scientific Research.

‡A large part of this work was completed while the author was at the National Magnet Laboratory.

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[7]It is safe to assume the oscillating electric field to be spatially homogeneous since the wavelength of microwave or infrared radiation is much larger than the cyclotron orbit radius and sometimes larger than the sample size.

[8]That the exact quantum mechanical solution of a driven harmonic oscillator takes this form is widely known. See, e.g., W. Louisell, Radiation and Noise in Quantum Electronics (McGraw-Hill Book Company, Inc., New York, 1964), p. 123. In this Letter, the emphasis, therefore, is on the exact solution of the specific physical problem rather than on the mathematical derivation.

[9]In the presence of a scattering mechanism (such as phonons), multiphoton resonances may be possible, though relatively small in amplitude.