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Ultrafast build-up of polarization in mesoscopic rings

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Abstract. – Subjecting a non-interacting mesoscopic ring (MR) to a linearly polarized half-cycle electromagnetic pulse results in a non-uniform, non-equilibrium coherent population of electronic states. A post-pulse charge polarization of the ring is then created within few picoseconds and sustains for times determined by the relaxation time scale. These conclusions are the results of a theoretical analysis of the electronic quantum dynamics of a MR subjected to unipolar pulses. Methods to trace experimentally the charge polarization build-up are suggested.

Introduction. – In mesoscopic systems the phase coherence length of the electrons can be considerably large compared to the system size. Hence, the physical properties of such systems are strongly influenced by the quantum interference of electronic states [1]. Prominent examples are the universal fluctuation of conductance, Anderson localization of electrons and the magnetic-field dependence of the thermodynamical and transport properties of multiple connected devices, *e.g.* mesoscopic rings (MRs) [1–5].

Extensive theoretical and experimental investigations (*e.g.* [1–9] and references therein) have established a common understanding of the equilibrium thermodynamics [1–4] as well as of the linear response of MRs to static or periodically time-dependent electric and magnetic fields [5, 6]. In contrast, the non-linear response of MRs is much less studied: *e.g.*, it has been shown [10–12] that a periodically time-dependent magnetic flux with a static component threading a MR generates a direct non-equilibrium current which is an odd function of the static magnetic-flux component (as is the case for persistent currents). Thus, the current vanishes in the absence of a static magnetic flux [11, 12]. The dynamics and the ac transport of MRs subject to an external cw laser have been studied in refs. [13, 14]. In this case, the charge polarization of an isolated MR, averaged over the period of the harmonic driving field, vanishes due to symmetry. The present paper provides a theoretical study of the dynamics of MRs under the action of short (on the typical electronic time scale of the ring) electromagnetic pulses. Such an investigation is most desirable for basically two reasons. On the one hand, as shown below, short unipolar or strongly asymmetric pulses have a qualitatively different influence on the dynamical properties of MRs than time-symmetric driving fields. Hence, truly new phenomena (*e.g.*, field-free charge polarization), occur when subjecting MRs to short, appropriately designed pulses. On the other hand, recently an enormous progress has been made in generating and shaping laser pulses [15]. It is conceivable that the use of the versatile possibilities of modern laser techniques to control and manipulate the dynamical properties of MRs will unveil a wealth of new phenomena. The present work is a first step

in this direction. Here we investigate the dynamics of a MR with non-interacting electrons subjected to a linearly polarized half-cycle pulse (HCP). An HCP is a strongly asymmetric mono-cycle pulse consisting of a very short, strong half-cycle (we refer to this part as an HCP), followed by a second half-cycle of an opposite polarity (the tail of the HCP) that can be substantially attenuated and stretched in time using optical gating techniques [16]. Since this tail is very weak and very long (compared to the relaxation time of MR) it hardly influences the electron dynamics. Nowadays, picosecond and subpicosecond HCPs with a peak field up to several hundred kV/cm [17] as well as trains of HCPs are experimentally available [18, 19].

The behavior of the electron dynamics in a MR when applying a cw laser or HCPs is markedly different. For a monochromatic cw laser no direct transfer of momentum to the electron takes place. In contrast, a short HCP acts on the electron as a directed impulsive *kick* (momentum change) given by $\boldsymbol{p} = -\int \boldsymbol{F}(t)\mathrm{d}t$, where $\boldsymbol{F}(t)$ is the time-dependent field of the HCP [16, 19, 20]. Therefore, the irradiation of the electronic system with HCPs may lead to qualitatively new phenomena. As an example, we demonstrate in the present work the existence of a field-free polarization of a MR following the application of a short HCP. More explicitly, an applied HCP with 1 ps duration delivers a directional momentum transfer to the electrons which results in post-pulse time-dependent charge density oscillations that last as long as the coherence is preserved. No current is induced in the ring because the applied HCP does not destroy the clockwise-anticlockwise symmetry of the charge carrier momenta in the ring. A further key difference between applying a cw laser and applying an HCP is that in case of a cw the fundamental laser frequency is the determining factor whereas a short HCP contains a multitude of frequencies. Hence, at first glance it may appear that an HCP contains a wide range of frequencies and can thus drive the system far from equilibrium. We remark however that in dependence of its initial state the system will respond only to certain frequencies of those offered by the pulse. Besides, the amplitudes of the HCP fields of interest here are small (less than 1 kV/cm) and the HCPs disturb the system only gently.

Theory. – We consider a two-dimensional, non-interacting MR at zero temperature exposed (at $t = t_1 = 0$) to an HCP which is linearly polarized along the x-direction. The applied pulses have a duration τ_d much shorter than the ballistic time τ_F which is the time lag a particle at the Fermi level (E_F) needs for completing one turn around the ring. This condition $\tau_\mathrm{d} \ll \tau_\mathrm{F}$ is currently feasible experimentally: for a typical ballistic ring τ_F is several tens of picoseconds [6, 9] and HCPs with $\tau_\mathrm{d} = 1$ ps are readily available [17]. The condition $\tau_\mathrm{d} \ll \tau_\mathrm{F}$ is important in that the interaction of the system with the HCP is then describable within the so-called impulsive (or sudden) approximation (IA) [19–21]. Physically, the IA means that an electron interacting with an HCP receives at the moment of irradiation an *instantaneous* directed momentum kick of the magnitude p (*i.e.* the time envelope $\varepsilon(t)$ of the pulse is approximated by a delta-function). For the results obtained below the IA is convincingly satisfied ($\tau_\mathrm{F} \gg \tau_\mathrm{d}$). The validity of the IA has been demonstrated numerically for a number of situations [20, 22] similar to the case considered here. Therefore, we present and analyze in this study the analytical predictions of the IA and illustrate them with some typical numerical examples. As mentioned above, we are concerned with clean ballistic rings. Hence the basic quantity which governs the quantum dynamics of the complete system is the single-particle, time-dependent wave function Ψ. The function Ψ is determined by solving for the time-dependent Schrödinger equation in the presence of the HCP, which in polar coordinates reads

$$i\hbar\frac{\partial\Psi}{\partial t} = \left[-\frac{\hbar^2}{2m^*}\left(\frac{\partial^2}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial}{\partial\rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial\theta^2}\right) + V_\mathrm{c}(\rho) - q\rho\varepsilon(t)\cos\theta\right]\Psi. \tag{1}$$

Here, the electron coordinate is specified by the radial position ρ and its angle θ measured with respect to the polarization vector of the HCP. The confining potential $V_{\rm c}(\rho)$ vanishes inside the ring and is infinitely large otherwise. The effective mass and the charge of the carriers are, respectively, m^* and q. The ring of interest for this study is such that $d \ll \rho_0$ where d and ρ_0 are, respectively, the ring's width and mean radius. The condition $d \ll \rho_0$ is well fulfilled for currently realized ballistic rings, *e.g.*, ref. [9] reported on ring fabrications with $d = 160\,\text{nm}$ and $\rho_0 = 1350\,\text{nm}$. In these thin rings the radial motion is much faster than the angular motion and, hence, the radial channels can be adiabatically decoupled from the angular motion. The stationary eigenstates of the system in the absence of the external field can then be written as $\Phi_{lm}(\rho,\theta) \approx R_l(\rho)\Theta_m(\theta)$, where the radial and angular components are given by

$$R_l(\rho) = \sqrt{\frac{2}{\rho_0 d}} \sin\left[\frac{l\pi}{d}(\rho - \rho_0 + d/2)\right], \qquad \Theta_m(\theta) = \frac{1}{\sqrt{2\pi}} e^{im\theta}. \tag{2}$$

The corresponding eigenenergies read $E_{lm} = \hbar^2(\pi^2 l^2 + m^2 d^2 \langle\rho^{-2}\rangle_l)/(2m^* d^2)$, where $\langle\rho^{-2}\rangle_l = \langle\Phi_{lm}(\rho,\theta)|\rho^{-2}|\Phi_{lm}(\rho,\theta)\rangle$, $l = 1, 2, 3, \ldots$, and $m = 0, \pm1, \pm2, \ldots$. The radial solution, given by eq. (2) is obtained upon neglecting the term $\propto (1/\rho)\partial/\partial\rho$ in the radial part of the Laplace operator. The validity of doing this for thin rings with $d \ll \rho_0$ has been discussed in ref. [23].

In what follows the wave function $\Psi_{l_0,m_0}(\rho,\theta,t)$ stands for the solution of eq. (1) with the initial (before applying the field) condition that the particle was residing in the electronic state characterized by the quantum numbers l_0 and m_0. For deriving $\Psi_{l_0,m_0}(\rho,\theta,t)$ we expand it in terms of the stationary eigenstates, *i.e.*

$$\Psi_{l_0,m_0}(\rho,\theta,t) = \sum_{l=1}^{\infty}\sum_{m=-\infty}^{\infty} C_{lm}(l_0,m_0,t)\Phi_{lm}(\rho,\theta)e^{-i\frac{E_{lm}t}{\hbar}}. \tag{3}$$

Inserting into eq. (1) we arrive, after some algebraic manipulation, at the following approximate expression for the expansion coefficients $C_{lm}(l_0,m_0,t)$:

$$C_{lm}(l_0,m_0,t) = \begin{cases} \delta_{l,l_0}\delta_{m,m_0}\,, & \text{for } t \le 0, \\ i^{m_0-m}J_{m-m_0}(\alpha_{l_0})\delta_{l,l_0}\,, & \text{for } t > 0, \end{cases} \tag{4}$$

where $\alpha_{l_0} = q\langle\rho\rangle_{l_0} p/\hbar$, $\langle\rho\rangle_{l_0} = \langle\Phi_{l_0 m}(\rho,\theta)|\rho|\Phi_{l_0 m}(\rho,\theta)\rangle$, and $J_l(x)$ are Bessel functions.

Upon applying the pulse the energy spectrum of the charge carriers is rearranged in a time-dependent manner. An analytical inspection of the time-dependent expectation value $E_{l_0 m_0}(t)$ of the Hamiltonian in the presence of the external field has led us to the following conclusion: within the IA the time-dependent energy of a particle occupying initially the $\{l_0, m_0\}$ state is given by

$$E_{l_0 m_0}(t) = \begin{cases} \dfrac{\hbar^2}{2m^* d^2}\left(\pi^2 l_0^2 + m_0^2 d^2 \langle\rho^{-2}\rangle_{l_0}\right), & \text{for } t \le 0, \\ \dfrac{\hbar^2}{2m^* d^2}\left[\pi^2 l_0^2 + \left(m_0^2 + \dfrac{\alpha_{l_0}^2}{2}\right) d^2 \langle\rho^{-2}\rangle_{l_0}\right], & \text{for } t > 0. \end{cases} \tag{5}$$

This relation evidences that an HCP shifts the unperturbed energy spectrum by an amount that scales quadratically with the kick strength p. p is finite and depends linearly on the HCP electric field amplitude. Hence, there is an upper bound for the energy an electron can achieve upon applying the pulse. The $\pm m_0$ initial state degeneracy is preserved after the

pulse is applied. Using eqs. (3)-(4) we deduce that $\Psi_{l_0,m_0}(\rho,\theta,t) = \Psi_{l_0,-m_0}(\rho,-\theta,t)$, *i.e.*, the clockwise-anticlockwise symmetry is preserved when applying an HCP. Therefore, currents carried by particles initially in the m_0 states are compensated for by the currents associated with the $-m_0$ states. From this fact and due to the degeneracy of the states (see eq. (5)), we conclude that no total current will circulate around the ring.

The expectation value of the induced dipole moment along the x-axis corresponding to the state $\{l_0, m_0\}$ is given by

$$\mu_{l_0,m_0}(t) = q\big\langle \Psi_{l_0,m_0}(\rho,\theta,t)\big|\rho\cos\theta\big|\Psi_{l_0,m_0}(\rho,\theta,t)\big\rangle. \tag{6}$$

From eqs. (3), (4), (6) an analytical expression for $\mu_{l_0,m_0}(t)$ can be derived, namely

$$\mu_{l_0,m_0}(t) = q\alpha_{l_0}\langle\rho\rangle_{l_0}\, Y(t)\, h\big(\Omega_{l_0}\big)\, \sin\left[\frac{2\pi t}{\tau_{l_0}}\right]\cos\left[\frac{4\pi m_0 t}{\tau_{l_0}}\right], \tag{7}$$

where $\Omega_{l_0} = \alpha_{l_0}\sqrt{2-2\cos[4\pi t/\tau_{l_0}]}$, $\tau_{l_0} = 4\pi m^*/(\hbar\langle\rho^{-2}\rangle_{l_0})$, and $Y(x)$ is the Heaviside step function. Furthermore, we introduced $h(\Omega_{l_0}) = J_0(\Omega_{l_0}) + J_2(\Omega_{l_0})$. From eq. (7) it is clear that $\mu_{l_0,m_0}(t) = \mu_{l_0,-m_0}(t)$. Thus, the contributions of electrons initially in the $\{l_0, m_0\}$ and $\{l_0, -m_0\}$ states to the polarization interfere constructively and a non-vanishing total polarization is generated.

The total HCP-induced dipole moment of the MR along the x-direction is then

$$\mu(t) = \sum_{l_0,m_0,\sigma} f\big(l_0, m_0, \sigma, N, T, t\big)\mu_{l_0,m_0}(t), \tag{8}$$

where σ refers to the charge carrier spin, N is the number of particles (which is assumed to be constant), T is the temperature and $\mu_{l_0,m_0}(t)$ is given by eq. (7). In eq. (8), the various μ_{l_0,m_0} have to be weighted with the non-equilibrium distribution function f. Under the present conditions f can be calculated within the relaxation time approximation [24] and eq. (8) takes on the form

$$\mu(t) = e^{-\frac{t}{\tau_{\rm rel}}}\sum_{l_0,m_0,\sigma} n_{\rm F}\big(l_0, m_0, \sigma, N, T, t\big)\ \mu_{l_0,m_0}(t), \tag{9}$$

where $\tau_{\rm rel}$ is the relaxation time and

$$n_{\rm F} = \left[1 + \exp\left[\frac{E_{l_0 m_0}(t>0) - \eta}{k_{\rm B}T}\right]\right]^{-1}, \tag{10}$$

with $k_{\rm B}$ and T representing the Boltzmann constant and the temperature, respectively. The value of the chemical potential η is calculated by requiring the number of particles in the ring to be a constant (recall we are considering an isolated ring). As shown by numerous investigations (*e.g.* [25] and references therein), the electron-electron interaction plays a minor role in determining the equilibrium properties of quasi–one-dimensional ballistic MRs containing a large number of carriers (as is the case in the present study). Therefore, the non-interacting electron model appropriately describes the equilibrium properties of such systems. Upon applying an HCP, however, the system is promoted to an excited non-equilibrium state and a polarization is induced in the ring. This polarization breaks the rotational symmetry and

leads to a violation of the momentum conservation. The situation is in close resemblance to what happened when the ring becomes "dirty", in which case the presence of disorder is responsible for breaking the rotational symmetry. Hence, the electron-electron scattering may become important for the relaxation process (to the equilibrium) of the HCP pumped system. Other relaxation mechanisms that can be relevant are the electron-phonon scattering, the simultaneous scattering by impurities and phonons, etc. A detailed study of these individual processes is interesting and probably feasible experimentally because, as will be shown below, the build-up time of the polarization can be varied by tuning the parameters of the exciting HCP. In the present work we focus on the presence of the polarization effect as such and hence account for the various relaxation processes by means of a single (averaged) quantity, namely the relaxation time $\tau_{\rm rel}$. It is worth remarking that as far as the ring is subject to a weak enough external field such that the system remains close to its equilibrium state, the non-interacting electron model is still expected to be a reasonable approximation. In this context we note that the ring here considered is only slightly disturbed by the HCP. *E.g.* for an electron with $E_{\rm F} = 4.228\,\text{meV}$, eq. (5) allows a maximum electron energy of $4.378\,\text{meV}$ following the irradiation with the 1 ps HCP with a peak amplitude of $F = 0.3\,\text{kV/cm}$ (cf. fig. 1 and the discussion below). Hence we do not expect a complete breakdown of the independent electron picture, valid for the initial state.

Numerical illustrations. – Explicit calculations were performed for a ballistic GaAs-AlGaAs–based MR of the type utilized in the experiment of ref. [9]. In such experimental ring the disorder was found to be very weak [9] and the "clean" ballistic ring model is evidently a good approximation. The relevant characteristics of the employed ring are $\rho_0 = 1350\,\text{nm}$, $d = 160\,\text{nm}$, the electron effective mass is $m^* = 0.067 m_{\rm e}$, and $N = 1400$. A typical HCP with a sine-square shape and a time duration of 1 ps has been used. At $T = 0\,\text{K}$, only the lowest-lying states are occupied and the total polarization is obtained by summing the contributions of all the radial channels. In case all the levels up to $E_{\rm F}$ are fully occupied (*i.e.*, if the number of particles in the ring obeys the relation $N = 2L + 4\sum_{l_0=1}^{L} M_{l_0}$) eq. (9) reads

$$\mu(t) = 2e^{-\frac{t}{\tau_{\rm rel}}} \sum_{l_0=1}^{L} \sum_{m_0=-M_{l_0}}^{M_{l_0}} \mu_{l_0,m_0}(t). \tag{11}$$

The number of channels L is defined as the largest integer $\leq k_{\rm F} d/\pi$ (*i.e.*, the largest channel index inside the Fermi surface) and M_{l_0} is the largest integer $\leq \sqrt{(k_{\rm F}^2 - (l_0^2\pi^2/d^2))/\langle \rho^{-2}\rangle_{l_0}}$ (*i.e.*, the largest occupied angular momentum in the l_0-th channel). The factor 2 in (11) accounts for the spin degeneracy. In the case of rings containing a large number of particles relation (11) is accurate, even when the highest occupied levels are not full.

Figures 1 (a) and (b) show the time dependence of the total dipole moment μ for different values of $\tau_{\rm rel}$ and for varying pulse amplitudes. We notice that a field-free *mesoscopic* polarization is generated within 30 ps after the application of the pulse (μ is shown in units of 10^6 debyes). The maximum absolute value of the post-pulse dipole moment decreases when shortening the relaxation time (see fig. 1 (a)). The post-pulse polarization, however, is still appreciable within a typical range of values of $\tau_{\rm rel}$ in ballistic semiconductor MRs as shown in fig. 1 (a). On the other hand, the dipole moment increases with the pulse strength but the time within which it is created decreases with stronger fields (see fig. 1 (b)). Thus, the amount and duration of the induced dipole moment can, to a certain extent, be tuned by applying an appropriately designed HCP. In principle, the post-pulse polarization and dipole moment can be enhanced considerably by increasing the field amplitude beyond the values

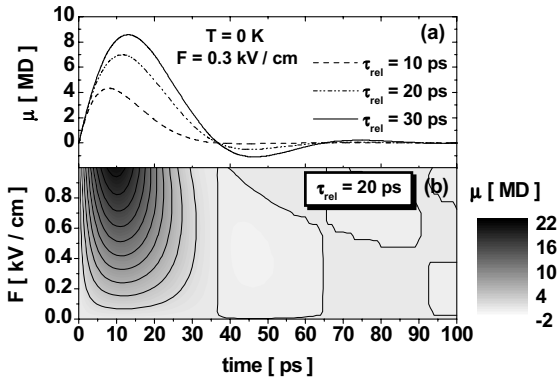

Fig. 1 – Time dependence of the total dipole moment $\mu(t)$ in units of 10^6 debyes as a function of the pulse field amplitude F. The pulse duration is 1 ps. In (b) the relaxation time $\tau_{\rm rel}$ is 20 ps. Inset (a) shows $\mu(t)$ at $F = 0.3\,\mathrm{kV/cm}$ and for different $\tau_{\rm rel}$. The calculations are at zero temperature.

shown in fig. 1. Stronger excitations result in shorter relaxation times and hence the post-pulses polarization and dipole moment will decay faster. In this context we emphasize that the charge polarization effect depicted in fig. 1 emerges after the HCP has passed by and hence it occurs in a nearly field-free environment. This fact offers a unique opportunity for studying relaxation processes in the absence of external perturbations and differs, qualitatively, from the case when a *stationary* polarization is induced by a dc electric field. It is worth noting that the polarization of the MR can be sustained for longer times if it is subject to a train of HCPs. For example, by applying a periodic train of HCPs (with a period longer than the relaxation time) the behavior of the polarization shown in fig. 1 can be periodically repeated as many times as the number of applied pulses. Due to the periodic charge oscillations, the driven MR (or arrays of MRs [8]) can result in a source of electromagnetic radiation whose characteristics can be controlled (to a certain extent) by appropriately designing the sequence of HCPs. In particular, the creation of a planar array of isolated MRs (similar arrays of connected MRs have already been experimentally realized [8]) is expected to resonantly increase the emission intensity [26].

In view of the size of the predicted effect and considering that all the parameters utilized in our investigation are in a range experimentally feasible with nowadays technology, it is of interest to point out ways to investigate experimentally the field-free charge polarization in MRs: Since the optical absorption properties depend strongly on the charge polarization state, the post-pulse charge polarization of the ring could be monitored by performing a pump-probe experiment analogous to that reported in ref. [27]. In our case, the HCP plays the role of the pumping beam that generates the ring charge polarization. A second delayed probe femtosecond pulse is then applied to monitor in time the absorption properties of the system (by varying the time delay between the pump and the probe fields). It is worth noting, however, that in contrast to ref. [27], in our case an HCP should serve as the pumping pulse. The predicted post-pulse charge polarization depends crucially on the amount of momentum transferred to the system by the pumping pulse. A time-symmetric, finite femtosecond pulse, as the one used in ref. [27], imparts no momentum change to the electrons and does not polarize the charge of the MR in the way described above. Another possibility for detecting the field-free polarization relies on inducing a current in the ring with a second HCP. If after a time delay Δt, a second HCP is applied (in a direction different from x breaking thus the clockwise-anticlockwise symmetry), a non-vanishing charge current is expected to be

induced in the ring. By monitoring the Δt-dependence of the current (or the induced ring magnetization) the dynamics of the post-pulse polarization can be revealed.

Conclusions. – In summary, we showed that the application of a linearly polarized HCP to a ballistic mesoscopic ring induces a post-pulse (and therefore field-free) time-dependent polarization. The time-dependent post-pulse polarization is expected to exist as long as the coherence is preserved and to decay in a time of the order of the relaxation time. Consequently, the field-free polarization effect could be useful for measuring relaxation times.

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