Magnetic flux quantization in dielectrics

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The oscillatory behavior of the magnetic moment is calculated as a function of the flux in a 1D Peierls dielectric with period doubling and also in a Peierls dielectric with an incommensurate charge density wave. The effect is comparable in magnitude to mesoscopic phenomena in normal-metal systems at ring dimensions smaller than or on the order of the correlation length.

One of the most interesting effects in which conduction electrons exhibit coherent properties in normal (i.e., nonsuperconducting) macroscopic systems is flux quantization. This effect, which was predicted back in 1970 by one of the present authors,[1] is a consequence of the Aharonov-Bohm effect. It consists of a periodic magnetic-flux dependence of thermodynamic and kinetic properties of hollow cylindrical conductors or of rings with a universal period equal to the normal flux quantum $\phi_0 = hc/e$. According to Ref. 1, the oscillation amplitude, which is quite sensitive to the temperature, is modulated by a factor of $\cos(k_F L + \alpha)$ (k_F is the Fermi wave vector of the electron. L is the perimeter of the sample, and α is a constant phase), which determines the mesoscopic (in today's terminology[2]) nature of the effect in the clean limit (see Refs. 3 and 4 regarding observation of this effect). In dirty systems, the mesoscopic oscillations with a period of hc/e are accompanied by oscillations in the kinetic properties with a period equal to half the flux quantum, $hc/2e$. These other oscillations are caused by weak localization effects.[5] Flux quantization was studied in Refs. 6 and 7 in semiconductors with a hopping conductivity mechanism. Finally, the existence of mesoscopic flux-quantization oscillations in a regime of 1D localization with a large relative amplitude (on the order of unity) was demonstrated in Ref. 8.

In all of the cases listed above, the flux quantization occurs in a system with free carriers, although the results of Ref. 8 also apply to so-called Anderson insulators. An important point is that it has been assumed everywhere that the filled states do not completely fill the Brillouin zone. In the present letter we wish to call attention to the possibility of flux quantization in dielectrics in which there are no free carriers. Specifically, we consider the ground-state energy of a dielectric with a periodic but otherwise arbitrary dispersion law

$$\epsilon(k) = \epsilon\left(k + \frac{2\pi}{a}\right), \tag{1}$$

where a is the size of the unit cell. In the field of the vector potential in the ring geometry (Fig. 1), we would have $A = \phi/L$. We then have the following expression for the total energy:

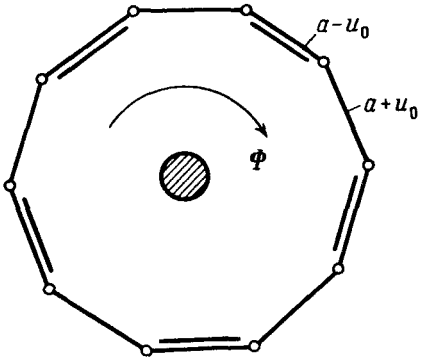

FIG. 1. Dielectric ring in the field of a vector potential.

$$W = \sum_{0 \leqslant k_n < 2\pi/a} \epsilon\left(k_n - \frac{e}{\hbar c} A\right), \qquad k_n = 2\pi n/L . \tag{2}$$

Expanding $\epsilon(k)$ in a Fourier series, we easily find

$$W = W_0 + \sum_s W_l \exp(-2\pi i l \Phi/\Phi_0) , \tag{3}$$

where W_0 is the energy of the filled band, which does not depend on ϕ, $l = sN$, N is the number of cells in the ring, and s is an integer. The reason for the higher harmonics in the dispersion law is a tunneling of electrons between remote neighbors; in an ordinary dielectric we would accordingly have $W_l \sim \exp(-sL/a)$, and at $L \gg a$ the flux quantization effect would be essentially unobservable. The situation changes if the number of higher harmonics in the dispersion law is large and if the small value of W_l is offset by this large number of harmonics.

An example of such a system is a Peierls dielectric, in which at low temperatures the electrons occupy a completely filled valence band, while the conduction band is empty and separated from the valence band by a gap of 2Δ. The quantity Δ is the modulus of the complex order parameter of the Peierls dielectric, which is equal to $\Delta e^{i\varphi}$. The phase φ of this order parameter describes the motion of a change density wave. The Hamiltonian of a Peierls dielectric is

$$H = \sum_{n,\sigma} t_{n,n+1} (a^+_{n\sigma} a_{n+1\sigma} e^{i\alpha_n} + \text{H.a.}) + \frac{\kappa}{2} \sum_n (u_n - u_{n+1})^2 , \tag{4}$$

where $t_{n,n+1} = t_0 + g(u_n - u_{n+1})$, u_n is the displacement of lattice site h, the operator $a^+_{u\sigma}$ creates an electron at site n with spin σ, $t_{n,n+1}$ is the electron transport integral, and κ is the elasticity of the lattice. In the simplest case of a Peierls dielectric with a dimerized lattice of the trans-$(CH)_x$ type, we would have $u_n = (-1)^n u_0$, there would be no phase degree of freedom; and the energy of the Peierls dielectric would be a function of exclusively the modulus $\Delta = 2gu_0$, where g is the electron-phonon coupling constant. Hamiltonian (4) can be diagonalized exactly; its spectrum

is ($\epsilon_F = 2|t_0|$)

$$\epsilon(k) = \pm \left\{ \Delta^2 + (\epsilon_F^2 - \Delta^2)\cos^2\left((k - \frac{e}{\hbar c} A)a\right) \right\}^{1/2} . \quad (5)$$

Correspondingly, the energy density of the ground state is written in the form

$$w = \frac{W}{L} = w_0 - \frac{\Delta}{2\pi\xi_0} + w_{osc} , \quad (6)$$

where $\xi_0 = \hbar v_F/\Delta$ is the coherence length of the Peierls dielectric. At $\epsilon_F \gg \Delta$ we have

$$w_{osc} \approx \frac{2\Delta}{\pi L} \sum_{s=1}^{\infty} \frac{1}{s} K_1(sL/\xi_0) \cos\left(2\pi s \frac{\phi}{\phi_0} - sk_F L\right) , \quad (7)$$

where K_1 is the modified Bessel function. Expression (7) contains both an oscillatory factor and a mesoscopic factor $\exp(isk_F L)$, where L is the perimeter. Both of these factors are typical of the flux quantization effect.[1] If the number of sides is even, the mesoscopic factor is equal to $(-1)^{Ns/2}$. If the number of sites is odd, a polaron excitation appears in the ring[9] and changes the magnitude of this factor. The value of Δ is found from the self-consistency condition for the Peierls dielectric and also contains an oscillatory component.

In macroscopic samples, $L \gg \xi_0$, it is sufficient to retain only the first harmonic in (7):

$$w_{osc} \approx \left(\frac{2}{\pi}\right)^{1/2} \frac{\Delta \xi_0^{1/2}}{L^{3/2}} e^{-L/\xi_0} \cos\left(2\pi \frac{\phi}{\phi_0} - k_F L\right). \quad (8)$$

We see that in place of the factor $\exp(-L/a)$ we have the vastly larger quantity $\exp(-L/\xi_0)$, reflecting the fact that there is a large number of high-index harmonics in dispersion law (5) for the Peierls dielectric.

Expression (7) was derived for $T = 0$, since the gap in a Peierls dielectric is usually large (on the order of 10^{3-4} K). On the other hand, if we formally take the limit $\Delta \to 0$, we find that (7) becomes the expression for normal metal, in which the harmonics decay in proportion to $\exp(-sTL/\hbar v_F)$.

In trans-$(CH)_x$ the coherence length is quite short, $\xi_0 \sim 10$ Å, and the effect can be observed only in rings of very small diameter ($\ll 100$ Å). However, the effect persists in Peierls dielectrics with large values $\xi_0 \sim 10^3$ Å (such as TaS_3, KO_3, MoO_3, $NbSe_3$; Ref. 10), in which the degree of commensurability is greater than twofold ($K_F a < \pi/2$). Specifically, the ground-state energy is the same as (6) in this case, as was shown in Ref. 9. There are grounds for believing that the part of the energy which is a periodic function of the flux will also fail to vanish in correlation dielectrics (i.e., Mott-Hubbard dielectrics) or in Peierls dielectrics in which the energy gap is formed as the result of a nesting of the Fermi surface and does not coincide with a boundary of the Brillouin zone in **k** space.

Oscillations in the thermodynamic properties are not the only consequences of

flux quantization. There are also certain other effects. For example, in a system with free carriers ($T \neq 0$) the conductivity of the system will oscillate. Since Δ is a nonmonotonic function of ϕ, the optical absorption near the threshold, $\hbar\omega \approx 2\Delta$, will also be a nonmonotonic function of the flux.

In a crystal made up of rings coupled by dispersion forces (of the benzene-molecule type, within which the binding is covalent), the magnetic susceptibility which follows from (7) is given in order of magnitude by the following expression under the condition $L < \xi_0$:

$$|\chi| \sim \frac{e^2}{\hbar v_F} \left(\frac{\epsilon_F}{\Delta}\right)^2 \left(\frac{v_F}{c}\right)^2 \sim 10^{-3} - 10^{-4}. \tag{9}$$

A general assertion which follows from the analysis above is of fundamental importance: The value of χ in dielectric crystals of this sort will oscillate as a function of an external magnetic field. However, the effect will be difficult to observe because of the large value of the period ($\Delta H \sim 10^7$ Oe at $L \sim 10$ Å).

In summary, it has been shown in this letter that the flux quantization effect occurs in dielectrics as well as conductors. For ordinary band dielectrics, the amplitudes of the oscillating harmonics in macroscopic rings are so small [$\sim \exp(-L/a)$] that the effect will be essentially unobservable. In dielectrics with a special spectrum containing many harmonics, the role of a will be assumed by a macroscopic quantity: a coherence length. If the dielectric state arises as a result of a localization of electrons (an Anderson transition), the amplitude will be determined by the relation between L and ζ, where ζ is the localization length of the electron states.

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