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Orbital diamagnetism of a charged Brownian particle undergoing a birth–death process

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Abstract. We consider the magnetic response of a charged Brownian particle undergoing a stochastic birth–death process. The latter simulates the electron-hole pair production and recombination in semiconductors. We obtain non-zero, orbital diamagnetism which can be large without violating the Van Leeuwen theorem.

1. Introduction

There is a well known theorem due originally to Van Leeuwen (1921) (see also Van Vleck 1932) on the absence of orbital diamagnetism in a classical system of charged particles in thermodynamic equilibrium. It is rigorous and holds for a completely general system Hamiltonian. The essential point of the proof (Pippard 1969, Peierls 1955) is that the magnetic field B enters the particle Hamiltonian only through the minimal replacement of the ith particle momentum $\boldsymbol{p}_i$ by $\boldsymbol{p}_i-(e/c)\boldsymbol{A}(r_i)$, where $\boldsymbol{A}$ is the associated vector potential. Now since the partition function involves integration of the particle momenta over the entire momentum space, the origin of which is trivially shifted by $\boldsymbol{A}$, the latter disappears from the partition function. This implies identically zero magnetic susceptibility. This partial tracing over the particle momenta is not permitted quantum mechanically because of the non-commutation problem. Here lies the origin of the Landau diamagnetism. For the purposes of this paper, two points about this classic theorem ought to be noted. Firstly, the proof of the theorem involves explicitly the system Hamiltonian which can, of course, be very general; and secondly, it makes no appeal to the thermodynamic limit—it holds for finite (bounded) systems. There is, however, a physically meaningful and formally well defined situation which is not covered by this theorem. This obtains when the charged particles in question are not permanent, in that they are being continually created and annihilated on a certain relevant timescale τ measuring the mean lifetime. An example would be a non-degenerate gas of electrons and holes suffering thermal pair production and recombination in a compensated semiconductor, or in a fluctuating valence system. In the language of chemical kinetics such a system may be termed 'reactive' (Nicolis and Prigogine 1977). Since there is no classical microscopic Hamiltonian describing such a process going on in the system in equilibrium, a description in terms of a partition function is inadmissible. One often resorts to a mesoscopic level of treatment based on

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some sort of master rate equation to compute a gross quantity such as the relative populations of the reacting species or their velocity distribution. In this paper we address ourselves to precisely such a system, idealised in some respects, and calculate its response to an external magnetic field of arbitrary strength. Our calculation shows that the orbital diamagnetic susceptibility is in general non-zero, and can indeed be very large for not too small but finite τ. In the limit $\tau \to \infty$ we do recover the result of the Van Leeuwen theorem, as we must. At the very outset, however, we must admit frankly that our enquiry at the moment is out of academic curiosity. We are unable to relate it to any definite experimental system, though we shall make some suggestions towards the end. On the other hand, we believe that our treatment, at the very least, throws considerable light on certain rather subtle questions relating to the role of boundary, or confinement, which is obscured in this theorem. This has some pedagogic value in itself. For a revealing discussion of these points in the quantum mechanical context, reference must be made to Pippard (1969).

2. Model and mathematical treatment

We shall follow here the natural description of the equilibrium statistical mechanics of such a non-Hamiltonian system in terms of the real space–time picture *à la* Langevin equation. In this picture the lifetime effect as stated above can easily be incorporated by superimposing on the particle history a stochastic birth–death process which is stationary in time.

Let us consider the simplest model of a classical system of charged particles, namely the Lorentz model in which we ignore all interactions among the particles but retain their interaction with any external field. It may be noted, however, that for a classical system of charged particles with the usual two-body interaction depending only on the inter-particle separation, the total magnetic moment of the system has a vanishing Poisson bracket with the two-body interaction part of the system Hamiltonian. This by itself implies that for such systems the two-body interactions do not alter the value of the magnetic moment, if there is a net magnetic moment at all. In our case the external field is a uniform magnetic field B directed along the positive z axis. The system will be assumed to be in thermal equilibrium with a bath (the lattice) at temperature T. It is sufficient, therefore, to examine the stochastic motion of just one 'test' particle moving under the influence of the randomly fluctuating bath forces and the external field. Further, for reasons of symmetry, we need consider only the motion projected onto a plane normal to the magnetic field, i.e. in the xy plane. In order to bring out certain points clearly, we shall first consider the case of permanent particles ($\tau = \infty$) moving in the infinitely extended xy plane, i.e. no boundary or confinement. We then have the Langevin equation (Chandrasekhar 1943)

$$m\ddot{x} = -\Gamma\dot{x} - \frac{|e|}{c}B\dot{y} + f_x(t), \qquad (1a)$$

$$m\ddot{y} = -\Gamma\dot{y} + \frac{|e|}{c}B\dot{x} + f_y(t), \qquad (1b)$$

where the random force field $f_\alpha(t)$ is a Gaussian white noise, i.e.

$$\langle f_\alpha(t) f_\beta(t')\rangle = A\delta_{\alpha\beta}\delta(t-t') \qquad (2a)$$

with $\alpha, \beta = x, y$. Γ is the frictional coefficient. The consistency condition for the state of equilibrium relates the prefactor A to Γ as

$$A = 2k_B T\Gamma. \tag{2b}$$

If we choose to look at the test particle in the course of its evolution at time t_0 ($<t$, the present epoch), it will have a certain velocity $\dot{x} = \dot{x}_0$, $\dot{y} = \dot{y}_0$ and position $x = x_0$, $y = y_0$, say. The condition (2b) ensures that asymptotically

$$\tfrac{1}{2}m\langle \dot{x}^2(t)\rangle_0 = \tfrac{1}{2}m\langle \dot{y}^2(t)\rangle_0 = \tfrac{1}{2}k_B T \tag{2c}$$

as $t \to \infty$, or equivalently, as $t_0 \to -\infty$. The latter implies that the system has been going on for an infinite time. The angular bracket $\langle \ldots \rangle_0$ denotes the subsemble average over the trajectories subject to the initial condition as noted above. This is merely an average over all possible trajectories of the particle starting from the given initial condition. Multiplying equation (1b) by i ($\equiv\sqrt{-1}$) and adding to equation (1a), we obtain

$$\ddot{z} = -\beta \dot{z} + F(t), \tag{3}$$

where $z = (x + iy)$, $F(t) = (1/m)(f_x(t) + if_y(t))$ and $\beta = (w_r - iw_c)$ with $w_r = \Gamma/m$ and $w_c = |e|B/mc$, the cyclotron frequency. We note that $z = x + iy$ occurs holomorphically in equation (3) and this simplifies the problem considerably. The quantity of interest is the magnetic moment

$$\langle m(t)\rangle_0 = \frac{|e|}{2c}\langle \boldsymbol{v} \times \boldsymbol{r}\rangle_0 \equiv -\frac{|e|}{2c}\,\mathrm{Im}\langle z^* \dot{z}\rangle_0 \tag{4}$$

evaluated in the limit $t_0 \to -\infty$. The formal solution of equation (3) is

$$z(t) = \frac{\dot{z}_0}{\beta}(1 - e^{-\beta(t-t_0)}) + \int_{t_0}^{t} d\eta\, e^{-\beta\eta} \int_{t_0}^{\eta} d\xi\, e^{\beta\xi} F(\xi), \tag{5}$$

where we have set $z_0 = 0$, which we can do without loss of generality. Recalling from equation (2a) that

$$\langle F(\xi)F^*(\xi')\rangle = 4k_B T\delta(\xi - \xi'), \tag{6}$$

we obtain

$$\begin{aligned}\langle m(t)\rangle_0 = -\Big(|\dot{z}_0|^2 \frac{|e|}{2c}\frac{1}{(w_r^2 + w_c^2)} e^{-w_r(t-t_0)}[w_r \sin w_c(t-t_0) \\ - w_c(\cos w_c(t-t_0) - e^{-w_r(t-t_0)})] + \frac{|e|}{mc}\frac{k_B T w_c}{(w_r^2 + w_c^2)} \\ \times (1 - e^{-2w_r(t-t_0)}) - \frac{2|e|}{mc}\frac{k_B T w_r}{(w_r^2 + w_c^2)} e^{-w_r(t-t_0)} \sin w_c(t-t_0)\Big).\end{aligned} \tag{7}$$

Now, letting $t_0 \to -\infty$, we should obtain the observable equilibrium value

$$\lim_{t_0 \to -\infty} \langle m(t)\rangle_0 = -\frac{|e|}{mc}\frac{k_B T w_c}{(w_r^2 + w_c^2)} \neq 0. \tag{8}$$

This non-zero result is manifestly in disagreement with the Van Leeuwen theorem and must, therefore, be wrong. This is in fact related to the well known paradox that in a real space–time picture an electron in a magnetic field must execute a cyclotron orbit and hence contribute a diamagnetic moment, whereas the above theorem predicts a zero value. The qualitative resolution of this paradox (see Pippard 1969) lies in the realisation that for a bounded system, the particle within an orbit diameter of the boundary must have its orbit intersect the boundary. This leads to skipping orbits in a sense counter to that of the cyclotron orbit of the particle in the bulk. This presumably gives exact cancellation. The statistical mechanical treatment of Van Leeuwen obscures this subtle role of the boundary which it subsumes. Indeed, as is well known, the mean-squared displacement $\langle |z(t)|^2 \rangle_0$ of our test particle grows as t for large t, even for non-zero B, and eventually as ergodicity demands, it must feel the effect of the boundary however remote (or equivalently, it must sense the confining potential however soft). Thus, even though the physically macroscopic systems are practically unbounded, we must notionally introduce a confining potential and only at the end let the potential strength tend to zero. We shall now demonstrate this explicitly by introducing a confining potential $\frac{1}{2}k(x^2+y^2)$, where the strength k is arbitrarily small, but non-zero to begin with. It turns out that we can again take $z_0 = 0$ without loss of generality. Now equation (3) is modified to

$$
\begin{aligned}
\langle m(t)\rangle_0 = -\frac{|e|}{2c}\,\mathrm{Im}\Bigg[&\frac{|\dot{z}_0|^2}{(\mu_1-\mu_2)(\mu_1^*-\mu_2^*)}\{\mu_1\exp[(\mu_1^*+\mu_1)(t-t_0)] \\
&+\mu_2\exp[(\mu_2^*+\mu_2)(t-t_0)]-\mu_2\exp[(\mu_1^*+\mu_2)(t-t_0)] \\
&-\mu_1\exp[(\mu_2^*+\mu_1)(t-t_0)]\}+\frac{f_0}{(\mu_1-\mu_2)(\mu_1^*-\mu_2^*)} \\
&\times\bigg(\frac{\mu_1}{(\mu_1^*+\mu_1)}\exp[(\mu_1^*+\mu_1)(t-t_0)]+\frac{\mu_2}{(\mu_2^*+\mu_2)}\exp[(\mu_2^*+\mu_2)(t-t_0)] \\
&-\frac{\mu_2}{(\mu_1^*+\mu_2)}\exp[(\mu_1^*+\mu_2)(t-t_0)]-\frac{\mu_1}{(\mu_2^*+\mu_1)}\exp[(\mu_2^*+\mu_1)(t-t_0)]\bigg)\Bigg],
\end{aligned} \tag{9}
$$

where

$$
w^2 = k/m, \qquad \mu_1 = -\tfrac{1}{2}\beta + (\beta^2/4 - w^2)^{1/2},
$$

$$
\mu_2 = -\tfrac{1}{2}\beta - (\beta^2/4 - w^2)^{1/2}, \qquad f_0 = (4k_B T w_r)/m.
$$

From equation (9) it can be verified, after some algebra, that $\langle m(t)\rangle_0$ indeed tends to zero as $t_0 \to -\infty$, in conformity with the Van Leeuwen theorem. In point of fact one verifies that the order of the limits $w \to 0$ and $t_0 \to -\infty$ is important, as anticipated earlier:

$$
\lim_{w\to 0}\Big(\lim_{t_0\to-\infty}\langle m(t)\rangle_0\Big) \to 0, \qquad \text{but} \lim_{t_0\to-\infty}\Big(\lim_{w\to 0}\langle m(t)\rangle_0\Big) \nrightarrow 0. \tag{10}
$$

This is an exact result. Earlier real space–time treatments of this exact cancellation were essentially heuristic and athermal—there was no bath. A few words of explanation are now in order to see qualitatively why the magnetic moment vanishes in the presence of the confining parabolic potential where the skipping cycles do not occur as such. The cancellation is due presumably to the fact that the restoring force of the confining potential causes the guiding centre of the Larmor orbit to move in a sense counter to that of the Larmor orbit itself. This can be qualitatively seen from equation (3) by including the restoring force due to the parabolic potential and dropping the damping and the concomitant fluctuating force terms. The resulting equation takes the form

$$\ddot{z} = \mathrm{i}w_c\dot{z} - kz,$$

which is an exactly solvable deterministic equation and displays the opposite directions of the motions of the guiding centre of the Larmor orbit and of the Larmor orbit itself.

Thus, reassured, we pursue this approach further to treat the case when τ is finite due to a stochastic birth–death process imposed on the particle history. In this case, however, it is not clear that we can set $z_0 = 0$ without loss of generality, and so we keep it general. The stochastic birth–death process is mathematically realised through a probability density $f(\boldsymbol{z}_0, \boldsymbol{v}_0; t-t_0)$ such that $f(\boldsymbol{z}_0, \boldsymbol{v}_0; t-t_0)\,\mathrm{d}\boldsymbol{z}_0\,\mathrm{d}\boldsymbol{v}_0\,\mathrm{d}t_0$ is the probability that the particle under observation at time t was 'born' in the space–time velocity element $\mathrm{d}\boldsymbol{z}_0\,\mathrm{d}\boldsymbol{v}_0\,\mathrm{d}t_0$. The observable magnetic moment $\langle m(t)\rangle$ is then

$$\langle m(t)\rangle = \int_{\boldsymbol{z}_0}\int_{\boldsymbol{v}_0}\int_{t_0=-\infty}^{t} f(\boldsymbol{z}_0, \boldsymbol{v}_0; t-t_0)\langle m(t)\rangle_0\,\mathrm{d}\boldsymbol{z}_0\,\mathrm{d}\boldsymbol{v}_0\,\mathrm{d}t_0. \tag{11}$$

Now, we make the reasonable assumption that at the time of birth 't_0' the particle is equally likely to be produced in all directions, i.e. the velocity distribution at birth is at least isotropic and hence $f(\boldsymbol{z}_0, \boldsymbol{v}_0; t-t_0)$ depends only on the magnitude v_0 of $\boldsymbol{v}_0$. This enables us to replace $\langle m(t)\rangle_0$ in equation (11) by its angular average $\langle\langle m(t)\rangle_0\rangle_{\mathrm{ang}}$ over the directions of $\boldsymbol{v}_0$. It can then be readily verified that $\langle\langle m(t)\rangle_0\rangle_{\mathrm{ang}}$ becomes independent of $\boldsymbol{z}_0$ and is given by equation (9) again. More explicitly, this is due to the fact that z_0 occurs in the expression for $\langle m(t)\rangle$ only through the combination $\langle \dot{z}_0 z_0\rangle$. Since, however, we have assumed the initial velocity distribution to be isotropic, the average $\langle \dot{z}_0 z_0\rangle$ vanishes by symmetry. Thus equation (11) becomes

$$\langle m(t)\rangle = \int_0^{\infty}\int_{-\infty}^{t} f_1(v_0; t-t_0)\langle\langle m(t)\rangle_0\rangle_{\mathrm{ang}}\,\mathrm{d}t_0\,\mathrm{d}v_0, \tag{12}$$

where

$$f_1(v_0; t-t_0) = \int_{\boldsymbol{z}_0}\int_{|\boldsymbol{v}_0|=v_0} f(\boldsymbol{z}_0, \boldsymbol{v}_0; t-t_0)\,\mathrm{d}\boldsymbol{z}_0\,\mathrm{d}\boldsymbol{v}_0. \tag{13}$$

Under very general conditions for the homogeneity of the stochastic process, we can make the Poissonian choice

$$f_1(v_0; t-t_0) = f_2(v_0)(1/\tau)\,\mathrm{e}^{-(t-t_0)/\tau}. \tag{14}$$

As is well known in the theory of stochastic processes any choice other than Poissonian necessarily implies memory. With this choice we obtain for the observed magnetic moment

$$\begin{aligned}\langle\overline{m(t)}\rangle = -\frac{|e|}{2c}\,\mathrm{Im}\Bigg\{&\frac{|\dot{z}_0|^2 w_0}{(\mu_1-\mu_2)(\mu_1^*-\mu_2^*)}\Bigg[\Bigg(\frac{\mu_2}{(\mu_1^*+\mu_2-w_0)}\\ &-\frac{\mu_1}{(\mu_1^*+\mu_1-w_0)}\Bigg)+(1\rightleftarrows 2)\Bigg]+\frac{f_0 w_0}{(\mu_1^*-\mu_2^*)(\mu_1-\mu_2)}\\ &\times\Bigg[\Bigg(\frac{\mu_1}{(\mu_2^*+\mu_1)(\mu_2^*+\mu_1-w_0)}-\frac{\mu_1}{(\mu_1^*+\mu_1)(\mu_1^*+\mu_1-w_0)}\Bigg)+(1\rightleftarrows 2)\Bigg]\Bigg\},\end{aligned} \tag{15}$$

where $\overline{|\dot{z}_0|^2}$ denotes initial velocity squared averaged over the distribution $f_2(v_0)$. One can readily verify that the equipartition requirement $\frac{1}{2}m\overline{|\dot{z}_0|^2} \to k_B T$ as $t \to -\infty$ gives $\overline{|\dot{z}_0|^2} = 2k_B T/m$. Equation (15) holds for arbitrary field strength so long as the demagnetisation effects are ignored.

First we note that in the limit of infinite lifetime, i.e. $\tau \equiv 1/w_0 \to \infty$, we recover the result that $\langle\overline{m(t)}\rangle \to 0$. With w_0 strictly greater than zero, we can let $w \to 0$, i.e. unbounded system, and obtain a much simpler expression for the orbital moment:

$$\langle\overline{m(t)}\rangle = -\frac{|e|}{2mc}\frac{k_B T w_c}{(w_r^2 + w_c^2)}\left(8 - \frac{4w_0}{(w_r + w_0)} - 16\frac{(w_0^2 + 2w_r w_0)}{(w_c^2 + (2w_0 + w_r)^2)}\right). \tag{16}$$

For very short lifetime, i.e. $w_0 \to \infty$, $\langle\overline{m(t)}\rangle$ tends to zero, which is physically quite understandable. For $0 < w_0 \ll w_r, w_c$, i.e. large but finite lifetime, the weak-field diamagnetic susceptibility χ per particle saturates at

$$\chi \text{ (per particle)} = -4\left(\frac{e}{mc}\right)^2 \frac{k_B T}{w_r^2}\frac{1}{(1 + w_0/w_r)^2}. \tag{17}$$

The limiting form in equation (17) can be given the following physical interpretation. As w_r increases, implying large frictional damping and concomitant fluctuating force, the motion of the particle becomes sufficiently randomised during the lifetime $1/w_0$ of the particle. The resulting loss of coherence presumably causes the reduction in the net magnetic moment as seen from equation (17), which has an overall dependence of the type $(w_r + w_0)^{-2}$. Expressing $w_r \equiv \Gamma/m$ in terms of the drift mobility μ by making use of the Einstein–Stokes relation, we obtain

$$\chi \text{ (per unit volume)} = -4n_0 \frac{k_B T \mu^2}{c^2}\frac{1}{(1 + w_0/w_r)^2}, \tag{18}$$

where n_0 is the number density of particles. For $n_0 \sim 10^{19}\ \text{cm}^{-3}$, $\mu \sim 10^3\ \text{cm}^2\ \text{V}^{-1}\ \text{s}^{-1}$ and $T \sim 300$ K, we obtain $\chi = 10^{-2}$ which is very large indeed.

3. Discussion

The foregoing treatment falls into two parts. The first part deals with the situation for which the Van Leeuwen theorem was originally envisaged. We are able to show explicitly the conceptually essential role played by the confinement in ensuring exact cancellation for which only heuristic arguments had been given so far. The second part is addressed to the case of a particle undergoing a birth–death process for which the theorem is not envisaged anyway. While it is gratifying that in the limit $\tau \to \infty$ we recover the known result, what is unphysical is that the relevant timescale $1/w_0$ is almost infinite for arbitrarily weak confinement (corresponding to the macroscopic size of the system in practice). In this sense the model is too ideal. We believe that in a real system the role of boundary is really played by the *bulk* imperfections like static inhomogeneities which are classically impenetrable and, of course, are not included in the Langevin equation. In such a case w may be effectively the reciprocal of the elastic mean free lifetime. If that is so, the condition for large diamagnetic susceptibility will be rather stringent, namely, $w < w_0 < w_r$. In indirect gap semiconductors, where electron-hole pair production–recombination requires phonons and hence depends sensitively on temperature and, of course, on compensation, the above condition may be realisable.

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