Temperature dependent diamagnetic–paramagnetic transitions in metal/semiconductor quantum rings

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We present theoretical studies of temperature dependent diamagnetic-paramagnetic transitions in thin quantum rings. Our studies show that the magnetic susceptibility of metal/semiconductor rings can exhibit multiple sign flips at intermediate and high temperatures depending on the number of conduction electrons in the ring (N) and whether or not spin effects are included. When the temperature is increased from absolute zero, the susceptibility begins to flip sign above a characteristic temperature that scales inversely with the number of electrons according to $N^{-1}$ or $N^{-1/2}$, depending on the presence of spin effects and the value of $N \mod 4$. Analytical results are derived for the susceptibility in the low and high temperature limits, explicitly showing the spin effects on the ring Curie constant.

I. INTRODUCTION

The electronic properties of low-dimensional structures with ring geometry have been a subject of great interest for many decades, starting with the early study of aromatic ring currents in benzene-like compounds [1] and later with persistent currents in microscopic conducting rings [2] [3]. Recent improvements in micro and nano-fabrication methods have renewed experimental interest in mesoscopic and quantum rings [4] [5], and in the past several decades numerous theoretical studies have continued to advance our understanding of these low dimensional metallic and semiconductor structures (see e.g. 4 [17]-23 and references therein). The use of quantum rings in real-world applications has also quickly advanced in relation to plasmonic devices and metamaterials [24] [25], which take advantage of the enhanced magnetic properties of nanorings by arranging them in precisely-controlled patterns. Effective design of these materials relies on predicting the magnetic susceptibility of the nanoring according to their shape, size, material, and temperature. However, as many others have pointed out [19] [27] [51], past estimates of the persistent current in conducting rings have differed from experimental measurements by orders of magnitude, and even predicting the sign of the low-field susceptibility has been problematic. Possible theoretical explanations for the discrepancies have included spin-breaking due to magnetic impurities [27], electron interactions [28], and experimental parameters like temperature or non-uniform probability of the number of electrons on the ring [30].

While earlier works have provided example calculations demonstrating the possibility of diamagnetic-paramagnetic transitions governed by temperature [30] [42], they do not provide a systematic study of this effect taking into account the influence of the rings size and material. Furthermore, these prior studies entirely disregard spin-induced Zeeman splitting of the energy levels, which we find can have a profound impact even for weak fields. In this paper we perform a systematic investigation of size and temperature effects on the ring susceptibility with and without spin effects. We find that as the temperature increases from absolute zero, the sign of the susceptibility can flip either once or multiple times, depending on the number of electrons on the ring (N) and whether or not spin effects are included. This diamagnetic-paramagnetic transition occurs above a certain critical transition temperature $T^*$ which decreases with the electron number according to either $N^{-1}$ or $N^{-1/2}$ power laws.

II. GRAND-CANONICAL APPROACH TO QUANTUM RINGS

We now consider the case of a quantum ring consisting of a fixed number $N$ of non-interacting electrons immersed in an external magnetic field. As has been well-established by now [22] [33] [36], it is most appropriate to use the canonical ensemble when the number of particles on the ring is fixed, but it is often a good approximation to treat the ring using a grand canonical ensemble and Fermi-Dirac statistics. Although there are known differences between the two approaches, we do not expect them to affect the qualitative trends discussed in this paper. Hence, in this work we present finite-temperature calculations in the grand canonical ensemble, and a more detailed discussion of the differences between the ensembles is reserved for a later work.

The thermodynamic properties of the system can be calculated using the grand canonical potential $\Omega$, which for fermions can be written as

$$\Omega (\mu, \kappa, T) = k_B T \sum_{m,\sigma} g_{m,\sigma} \ln \left[ 1 - f_{m,\sigma} (\mu, \kappa, T) \right] \quad (1)$$

where $f_{m,\sigma} (\mu, \kappa, T) = (1 + \exp [(\epsilon_{m,\sigma} - \mu)/k_B T])^{-1}$ is the Fermi-Dirac distribution, and $\epsilon_{m,\sigma}$ and $g_{m,\sigma}$ are the energy levels of the system and their respective degeneracy factors. The chemical potential $\mu$ is obtained from the thermodynamic average of the system total electron number

$$N = - \frac{\partial \Omega}{\partial \mu} = \sum_{m,\sigma} g_{m,\sigma} f_{m,\sigma} (\mu, \kappa, T) . \quad (2)$$
The energy levels $\epsilon_{m,\sigma_z}$ are found from the Schrödinger–Pauli equation $\hat{H}\psi_{m,\sigma_z} = \epsilon_{m,\sigma_z}\psi_{m,\sigma_z}$. For a one-dimensional (1D) quantum ring with radius $R$ immersed in a constant magnetic field $B = B\hat{z}$ perpendicular to the plane of the ring, the Hamiltonian reads as $\hat{H} = \Delta \left(-i\partial_\phi + \kappa \right)^2 + g_s\sigma_z\kappa$, where $\Delta = \hbar^2/2m_z R^2$ is the zero-field energy spacing between the ground level and the next-highest level, and $\kappa = \Phi/\Phi_0$ is a flux parameter written in terms of the magnetic flux through the ring $\Phi = \pi R^2 B$ and the flux quantum $\Phi_0 = 2\pi\hbar/e$.

In this study we also account for the interaction of the conduction electrons then follow as

$$\psi_{m,\sigma_z} = \frac{1}{\sqrt{2\pi R}} \chi_{\sigma_z} e^{im\phi},$$

$$\epsilon_{m,\sigma_z}(\kappa) = \Delta \left((m + \kappa)^2 + g_s\sigma_z\kappa\right), \tag{3}$$

where $m = 0, \pm 1, \ldots$ is the azimuthal quantum number and $\chi_{\sigma_z}$ is the spin part of the wavefunction. Because we consider classical free electrons, we can take $g_s = 2$ to a high degree of accuracy, but retaining the factor as a parameter has a couple of advantages. First, it allows the model to be extended naturally to some semiconductor materials in which the effective Landé factor differs from that of free electrons. A second advantage is that in the model we can switch on or off the Zeeman splitting effect as needed. This is important in the study of persistent currents and Aharonov-Bohm rings where the magnetic field is presumed to only penetrate the interior of the ring. Thus, the two cases $g_s = 0$ and $g_s = 2$ represent the two extremes of spin effects turned on or off.

Once the grand canonical potential is obtained we find the magnetic susceptibility of the ring which follows from thermodynamic considerations as

$$\chi = -\mu_0 V \left(\frac{\partial^2 \Omega}{\partial B^2}\right)_{T,\mu} \bigg|_{B=0},$$

$$\epsilon_{m,\sigma_z}(\kappa) = \Delta \left((m + \kappa)^2 + g_s\sigma_z\kappa\right), \tag{4}$$

where $\mu_0 = \mu_0 B$.

Applying Eq. (2) to Eq. (1) with the energy levels in Eq. (1) and noting that the degeneracy due to Zeeman splitting of the energy levels is $g_{m,\sigma_z} = 1$ if $g_s \neq 0$ and $g_{m,\sigma_z} = 2$ if $g_s = 0$, we obtain

$$\chi = \left|\chi_L\right| \left(-1 + \frac{16T_F}{N^4 \Omega} \sum_{m=-\infty}^{\infty} \left(m^2 + g_s^2 \right) \right) \times \text{sech}^2 \left(\frac{8T_F}{N^2 \Omega} (m^2 - \bar{\mu}(0))\right), \tag{5}$$

where the Langevin susceptibility due to Larmor precession is $\chi_L = -N\pi R^2 \alpha^2 a_B/\hbar = -N\chi_1$ where $a_B$ denotes the Bohr radius and $\alpha$ is the fine-structure constant. In writing Eq. (3) we have used tilde notation to indicate the dimensionless chemical potential at zero field $\bar{\mu}(0) = \mu(\kappa = 0)/\Delta$ and defined the ring 1D Fermi temperature

$$\epsilon_F = k_B T_F = \Delta \left(\frac{N}{4}\right)^2 \tag{6}$$

with $k_B$ Boltzmann's constant. Note that this one-dimensional Fermi temperature follows from evaluating Eq. (2) in the limit $N \to \infty$ and should not be confused with the Fermi temperature of bulk material. We write our results in terms of this Fermi temperature since it can be applied to real materials in a phenomenological way by relating the radius of the ring to the number of particles through $N = \pi R^2 r_s$ where $r_s = (3/\left(4\pi n_e \right))^{1/3}$ is the Wigner-Seitz radius and $n_e$ is the free-electron density. Inserting this relation in Eq. (6) leads to $T_F = \pi^2 \hbar^2 / (32 m_e k_B r_s^2)$. For example if we take $r_s = 1.60 \text{ Å}$ for silver we find $T_F = 10,690 \text{ K}$.

### III. Ring Size and Temperature Dependence of the Susceptibility

Here we study the characteristics of the susceptibility as a function of the temperature and the total number of conduction electrons $N$. Figure 1 shows Eq. (5) evaluated for some exemplary cases at different fixed temperatures, with and without spin effects included. The results show four distinct cases for the susceptibility depending on the value of $N \mod 4$, which we label in Fig. 1 using the integer $j = 0, 1, 2, \ldots$. These four cases correspond to the possible number of paired spin-1/2 particles following the Pauli exclusion principle. This well-known $N \mod 4$ property of one-dimensional rings is sometimes referred to as a double-parity effect [32, 37]. As seen in Fig. 1(a), all $N = 4j + 2$ cases show diamagnetic behavior for small $N$. This phenomenon is known as Hückel's rule in the context of aromatic chemistry [33, 39] and represents the case where all electron spins are paired. By contrast, the other set of even-numbered rings ($N = 4j + 4$) are paramagnetic, similar to odd-numbered rings at the chosen temperatures. In the case of a Hückel type ring the magnetic susceptibility is found to follow the Langevin susceptibility $\chi = \chi_L$ until reaching $N \approx T_F/T$ and then decays with increasing $N$ (see Fig. 1(a)). The maximum diamagnetic response can also be estimated at $\chi_{\max} \approx -\chi_1 T_F/T$. If $h$ is the physical thickness of the ring then $\chi_1 = -\alpha^2 a_B/h$. For any physical conducting ring we have $h \geq a_B$, and hence the maximum diamagnetic susceptibility is given by the rather simple result $\chi_{\max} \approx -N\alpha^2$. We also see that spin has a significant impact compared to orbital effects when $N \gg T/T_F$. For fixed temperature and increasing $N$, the susceptibility
Figure 1. The magnetic susceptibility with spin (dashed lines) and without spin (solid lines), shown at two fixed temperatures, $T/T_F = 0.015$ (black) and $T/T_F = 0.03$ (red). **Even number of electrons:** (a) $N = 4j + 2$ (Hückel) and (b) $N = 4j + 4$. **Odd number of electrons:** (c) $N = 4j + 1$ and (d) $N = 4j + 3$. Arrows indicate the regions in which the susceptibility flips sign when spin effects are present. Blue dotted lines indicate the limits for $N \ll T_F/T$ and $N \gg T_F/T$ when spin is included.

Decays exponentially if there is no spin and decays with a power law $1/N$ when spin is present. In the $N = 4j + 1$ and $N = 4j + 2$ cases, when $N \to \infty$ the sign of the susceptibility depends on whether spin effects are included. For large $N$, the $N = 4j + 2$ rings are diamagnetic without spin included, but when spin is included, they transition to paramagnetism above a critical size. The limiting behavior for both small and large $N$, shown as blue dotted lines in Fig. 1, follow from the low and high temperature limits derived in the sections that follow.

Finally, in Fig. 1 we also observe a strong temperature dependence with the magnetic susceptibility increasing in absolute values as the temperature is lowered. With this in mind, we now turn to the primary focus of this paper, which is to better understand the susceptibility across a broad range of temperatures. In Fig. 2, we find that at high temperatures the susceptibility decays exponentially without spin but decays more slowly when spin is present, acquiring a nearly constant value for $T \ll T_F$. The visible kinks in the logarithmic plots reveal that the susceptibility can flip sign at intermediate temperatures. These results demonstrate that there are three recognizable temperature regimes: low temperature, high temperature, and intermediate temperature. We further analyze each regime separately in the following three sections.

A. Low temperature limit

When $T \ll T_F/N$, the Fermi-Dirac function $f_{m,\sigma_z}(\mu, \kappa, T)$ acts like a step function, and only the $N$ lowest energy levels are occupied. In this low-temperature limit, the chemical potential depends sen-
Figure 2. The temperature dependence of the magnetic susceptibility with spin (dashed lines) and without spin (solid lines). **Even number of electrons:** (a) \( N = 4j + 2 \) (Hückel) and (b) \( N = 4j + 4 \). **Odd number of electrons:** (c) \( N = 4j + 1 \) and (d) \( N = 4j + 3 \). Arrows indicate the regions in which the susceptibility flips sign when spin effects are included. Blue dotted lines indicate the limits for \( T \gg T_F/N \) (when spin is included) and \( T \ll T_F/N \) (with or without spin).

Even number of electrons:

\[
N = 4j + 2 \text{(Hückel)} \quad \text{and} \quad N = 4j + 4.
\]

Odd number of electrons:

\[
N = 4j + 1 \quad \text{and} \quad N = 4j + 3.
\]

The results of Eq. (9) are shown as blue dotted lines at low temperatures in Fig. 2, displaying the \( 1/T \) behavior in the cases \( N = 4j + 1, 4j + 3, \) and \( 4j + 4 \).
B. High temperature limit

We observed in Fig. 2 that for all cases of $N$ the susceptibility decays rapidly with increasing temperature. When spin is not included, the susceptibility decays exponentially and experiences an infinite number of sign flips. When spin is included, the susceptibility decays more slowly and either remains paramagnetic or experiences a diamagnetic-paramagnetic transition before approaching zero. This important finding predicts that all rings will display a paramagnetic response at sufficiently high temperatures when spin effects are present. To see why this is the case, we proceed with an analytical evaluation of Eq. (5) at high temperatures. When the thermal energy is much greater than the energy-level spacing at the Fermi surface ($k_B T \gg \epsilon_F/N$) or equivalently, $T \gg T_F/N$, the energy levels form a nearly continuous band, and the summations in Eqs. (2) and (5) can be approximated using integration by applying the Euler-Maclaurin formula (see Appendix for details). The resulting integrals can be evaluated in closed-form using special functions, finding for the chemical potential

$$\mu(0) = \frac{N^2 T}{16 T_F} \log \left( -\text{Li}_{1/2}^{-1} \left( -2 \sqrt{\frac{T_F}{\pi T}} \right) \right)$$

and the susceptibility

$$\chi_m = \frac{4g_s^2 |\chi_L|}{N^2} \sqrt{\frac{T_F}{T}} \text{Li}_{1/2}^{-1} \left( \text{Li}_{1/2}^{-1} \left( -2 \sqrt{\frac{T_F}{\pi T}} \right) \right)$$

where $\text{Li}_n(x)$ are the polylogarithm functions of order $n$ and $\text{Li}_{1/2}^{-1}(x)$ denotes the inverse of the polylogarithm. Making use of the asymptotic values $\text{Li}_{1/2}^{-1} \left( -2 \sqrt{T_F/(\pi T)} \right) \approx -e^{T_F/T}$ and $\text{Li}_{1/2}^{-1} \left( -e^{T_F/T} \right) \approx \sqrt{T/(\pi T_F)}$ in the limit $T \ll T_F$, we find from Eq. (11)

$$\chi_m = \frac{4g_s^2 |\chi_L|}{N^2} \frac{T_F}{N} \ll T_F.$$  

Thus at high temperatures with spin effects included ($g_s \neq 0$), a paramagnetic susceptibility is always expected regardless of $N \mod 4$ (see blue dotted lines on the right sides of Fig. 2). Note that this high temperature limit also serves as the bulk limit in Fig. 1 where the criterion $T \gg T_F/N$ is satisfied for large $N$. This paramagnetism in the bulk and high temperature limits is purely a spin effect since the magnitude of the susceptibility decays to zero exponentially with increasing temperature when spin is absent.

C. Paramagnetic-diamagnetic transitions

We have already seen in Fig. 2 that the sign of the susceptibility can experience single or multiple flips at intermediate temperature values when $T \approx T_F/N$. Figure 3 further visualizes how the transitions between diamagnetism and paramagnetism depend on the number of particles, the temperature, and whether or not spin is included. The diamagnetic Hückel rings with $N = 4j + 2$ are clearly visible as dark blue bars at the bottom of the figure where $T \ll T_F/N$. The other three cases for $N$ show strong paramagnetism at low temperatures, shown as dark red bars. As the temperature increases, the susceptibility exponentially decays as it approaches zero in all cases. However, the result of including spin effects becomes apparent with comparing the left and right panels of Fig. 3. From the left panel we see that if only angular momentum plays a role (no spin effects) the susceptibility experiences rapid change of sign as the temperature is increased. On the other hand, in the right panel we observe that when spin effects are included and the rings number of electrons corresponds to Hückel rule ($N = 4j + 2$) the susceptibility flips only once from diamagnetic to paramagnetic. For rings with $N = 4j + 1$ the susceptibility flips exactly twice (for $N \geq 13$) while for all other cases it remains paramagnetic. Furthermore, in accordance to our high temperature analysis (see Eq. (11) and discussions thereafter), all cases manifest a paramagnetic response for sufficiently high temperatures. Inspecting Figs. 2 and 3, there is clearly a size-dependent transition temperature beyond which paramagnetic-diamagnetic transitions are manifested. We track the size-dependence of this transition by defining the temperature $T^*$ to be the lowest temperature at which the susceptibility changes sign. This transition temperature is presented in Fig. 4 where calculations have been performed for each case of $N \mod 4$ with $g_s = 0$ in Fig. 4(a) and $g_s = 2$ in Fig. 4(b). The $N = 4j + 3$ and $N = 4j + 4$ rings are not shown in Fig. 4(b) since they never change sign when $g_s = 2$. The dashed line in Fig. 4(b) indicates the second flip of $N = 4j + 1$ rings so the region between the dashed and solid red lines indicates the range of temperatures in which $N = 4j + 1$ rings exhibit diamagnetism.

We find that $T^*$ decreases rapidly as the number of electrons increases. In particular, the logarithmic plot shows that this temperature follows a size-dependent power law $T^* \sim N^{p(N)}$, where the exponent $p(N)$, shown in the bottom panels of Fig. 4, corresponds to the slope of the lines in the top panels. For all cases we have $-1 \leq p(N) \leq -1/2$. Denoting by $p_\infty$ the asymptotic behavior in the bulk limit $N \to \infty$, in the case when spin effects are not included we obtain from numerical convergence the values of the exponents as

$$p_\infty = \begin{cases} -1, & N = 4j + 1, \\ -1/2, & \text{else} \end{cases}, \quad g_s = 0. \tag{13}$$

If spin is included, we obtain

$$p_\infty = \begin{cases} -1, & N = 4j + 1, \\ -1, & N = 4j + 2, \quad g_s = 2. \end{cases} \tag{14}$$

Note that the difference in $p_\infty$ between the cases with and without spin can drastically change the temperature at which diamagnetic-paramagnetic transitions appear. This strong discrepancy could potentially serve
Figure 3. The susceptibility as the temperature and number of electrons are varied, shown (left) without spin effects and (right) with spin effects. Blue regions indicate diamagnetic susceptibility and red regions are paramagnetic.

IV. CONCLUSION

We have investigated the temperature dependence of the magnetic susceptibility in one-dimensional conductive rings for a wide range of temperatures and sizes. Analytical results were provided for the low and high temperature limits. At low temperatures $T \ll T_F/N$, conducting rings with a total number of conduction electrons corresponding to Hückels rule ($N = 4j + 2$) are always diamagnetic, reaching a maximum value $\chi_{\text{max}} \approx -\chi_1 T_F/T$. For non-Hückel rings, a Curie-type paramagnetism is observed. At high temperatures $T \gg T_F/N$ and for all values of $N$, the susceptibility exponentially decays with increasing temperature, approaching $\chi \to 0$ when spin effects are absent. When spin is included, the susceptibility becomes weakly paramagnetic at high temperatures, acquiring the value $\chi = 4g_s^2/N^2$ for $T_F/N \ll T \ll T_F$. For intermediate temperatures higher than a critical value $T^*$, our studies show that the conductive ring susceptibility experiences complex behavior including single or multiple diamagnetic-paramagnetic transitions. The bulk limiting behavior of this critical temperature is either $T^* \sim N^{-1/2}$ or $T^* \sim N^{-1}$ depending on $N \mod 4$ and the presence of spin effects. The predicted magnetic transitions should be subject to experimental studies since data for isolated individual rings is still lacking.

Appendix A: Chemical potential in the high temperature limit

In this appendix, we provide details of the calculation of the high-temperature result for the chemical potential. Written explicitly using Eqs. (2) and (3), the chemical potential is defined by

$$N = \sum_{m=-\infty}^{\infty} \left( \frac{1}{1 + e^{\frac{\Delta}{k_B T}}} \left( (m+\kappa)^2 + g_s \kappa - \tilde{\mu} \right) \right) + \frac{1}{1 + e^{\frac{\Delta}{k_B T}}} \left( (m+\kappa)^2 - g_s \kappa - \tilde{\mu} \right).$$
Changing the limits of the summation, we find the equivalent form

\[ N = -\frac{f(0)}{2} + \sum_{m=0}^{\infty} f(m) \]  

(A1)

where

\[ f(m) = \frac{1}{1 + e^{\frac{\Delta}{k_BT}((m+\kappa)^2 + g_s \kappa - \bar{\mu})}} + \frac{1}{1 + e^{\frac{\Delta}{k_BT}((m+\kappa)^2 - g_s \kappa - \bar{\mu})}} + \frac{1}{1 + e^{\frac{\Delta}{k_BT}((-m+\kappa)^2 + g_s \kappa - \bar{\mu})}} + \frac{1}{1 + e^{\frac{\Delta}{k_BT}((-m+\kappa)^2 - g_s \kappa - \bar{\mu})}}. \]

Applying the Euler-Maclaurin formula, we find

\[ \sum_{m=0}^{\infty} f(m) = \int_0^\infty f(m) \, dm + f(0)/2 \]

where we have used that \( f^{(2k-1)}(0) = f^{(2k-1)}(\infty) = f(\infty) = 0 \). With this result and Eq. (A1), we can now write

\[ N = \int_0^\infty f(m) \, dm. \]

With the substitutions \( x = m + \kappa \) and \( y = m - \kappa \), we can split the integral in the following way

\[
\int_0^\infty f(m) \, dm = \int_0^\infty F(x) \, dx - \int_0^{\kappa} F(x) \, dx + \int_0^\infty G(y) \, dy - \int_0^{\kappa} G(y) \, dy
\]

where the integrands are defined by

\[
F(x) = \frac{1}{1 + e^{\frac{\Delta}{k_BT}(x^2 + g_s \kappa - \bar{\mu})}} + \frac{1}{1 + e^{\frac{\Delta}{k_BT}(x^2 - g_s \kappa - \bar{\mu})}}
\]

\[
G(y) = \frac{1}{1 + e^{\frac{\Delta}{k_BT}(y^2 + g_s \kappa - \bar{\mu})}} + \frac{1}{1 + e^{\frac{\Delta}{k_BT}(y^2 - g_s \kappa - \bar{\mu})}}.
\]

Recognizing that \( x \) and \( y \) are dummy integration variables, we have

\[
\int_0^\infty F(x) \, dx = -\int_0^{\kappa} G(y) \, dy
\]

\[
\int_0^{\infty} F(x) \, dx = \int_0^{\infty} G(y) \, dy
\]

so we are left with \( \int_0^{\infty} f(m) \, dm = 2 \int_0^{\infty} F(x) \, dx \), giving

\[
N = 2 \int_0^{\infty} \left( \frac{1}{1 + e^{\frac{\Delta}{k_BT}(x^2 + g_s \kappa - \bar{\mu})}} + \frac{1}{1 + e^{\frac{\Delta}{k_BT}(x^2 - g_s \kappa - \bar{\mu})}} \right) \, dx.
\]

This integral can be evaluated using polylogarithm functions, leading to a transcendental equation for \( \bar{\mu} \),

\[
N = -\sqrt{\frac{\pi k_B T}{\Delta}} \left( \text{Li}_{\frac{1}{2}} \left( -e^{\frac{\Delta}{k_BT}(-\bar{\mu} - g_s \kappa)} \right) + \text{Li}_{\frac{1}{2}} \left( -e^{\frac{\Delta}{k_BT}(\bar{\mu} + g_s \kappa)} \right) \right)
\]

(A2)

where \( \text{Li}_n(x) \) are the polylogarithm functions of order \( n \). Evaluating Eq. (A2) at \( \kappa = 0 \) gives

\[
N = -2 \sqrt{\frac{\pi k_B T}{\Delta}} \text{Li}_{\frac{1}{2}} \left( -e^{\frac{\Delta(\bar{\mu})}{k_BT}} \right),
\]
and substituting $T_F$ for $\Delta$ via Eq. [6], we arrive at the result
\[
-1 = \frac{\sqrt{\pi}}{2} \left( \frac{T}{T_F} \right)^{1/2} \text{Li}_{\frac{1}{2}} \left( -e^{\frac{16T_F\tilde{\mu}(0)}{N^2}} \right) \quad (A3)
\]
Finally we find Eq. [10] when we solve Eq. [A3] for the chemical potential. Making use of the asymptotic value $-\text{Li}_{\frac{1}{2}} \left( -2\sqrt{T_F/(\pi T)} \right) \approx e^{T_F}$ for $T \ll T_F$ recovers the bulk result $\tilde{\mu} (0) = N^2/16$ quoted in the text.

**Appendix B: Susceptibility in the high temperature limit**

Here we derive the high-temperature susceptibility result given by Eq. [11] in the text. We begin by changing the limits of the summation in Eq. [5], finding the equivalent form
\[
\chi_m = |\chi_L| \left( -1 + \frac{2b}{N} \sum_{m=0}^{\infty} f(m) \right)
= |\chi_L| \left( -1 + \frac{4b}{N} \left( f(0)/2 + \sum_{m=0}^{\infty} f(m) \right) \right)
\]
where $b = 8T_F/(N^2T)$ and
\[
f(m) = \left( m^2 + \frac{g_0^2}{4} \right) \text{sech}^2 \left( b \left( m^2 - \tilde{\mu}(0) \right) \right)
\]
Now applying the Euler-Maclaurin formula in the same way as we did for the chemical potential in Appendix A, we find $\sum_{m=0}^{\infty} f(m) = \int_0^{\infty} f(m) \, dm + f(0)/2$ since again we have $f^{(2k-1)}(0) = f^{(2k-1)}(\infty) = f(\infty)$. This leads to
\[
\chi_m = |\chi_L| \left( -1 + \frac{4b}{N} \int_0^{\infty} f(m) \, dm \right), \quad (B1)
\]
and with the substitution $x = bm^2$ the integral becomes
\[
\int_0^{\infty} f(m) \, dm = \frac{1}{2b^{3/2}} \int_0^{\infty} x^{1/2} \text{sech}^2 \left( x - b\tilde{\mu}(0) \right) \, dx
+ \frac{g_0^2}{8b^{1/2}} \int_0^{\infty} x^{-1/2} \text{sech}^2 \left( x - b\tilde{\mu}(0) \right) \, dx.
\]
With the help of the integral formula
\[
\int_0^{\infty} x^n \text{sech}^2 (x-a) \, dx = -2^{1-n} \Gamma (n+1) \text{Li}_n \left( -e^{2a} \right)
\]
valid for $n > -1$, we find
\[
\int_0^{\infty} f(m) \, dm = -\frac{1}{2} \sqrt{\frac{\pi}{b^{3/2}}} \text{Li}_{\frac{1}{2}} \left( -e^{2b\tilde{\mu}(0)} \right)
+ \frac{g_0^2}{b^{1/2}} \text{Li}_{-\frac{1}{2}} \left( -e^{2b\tilde{\mu}(0)} \right).
\]
With this result, Eq. [B1] becomes
\[
\chi_m = |\chi_L| \left( -1 - \sqrt{\frac{\pi}{b}} \left( \frac{T}{T_F} \right)^{1/2} \text{Li}_{\frac{1}{2}} \left( -e^{\frac{16T_F\tilde{\mu}(0)}{N^2T}} \right) \right)
- \frac{4g_0^2\sqrt{\pi}}{N^2} \left( \frac{T}{T_F} \right)^{-1/2} \text{Li}_{-\frac{1}{2}} \left( -e^{\frac{16T_F\tilde{\mu}(0)}{N^2T}} \right). \quad (B2)
\]
Inserting the relationship Eq. [A3] into Eq. [B2], we find
\[
\chi_m = -\frac{4g_0^2\sqrt{\pi}}{N^2} \left( \frac{T}{T_F} \right)^{-1/2} \text{Li}_{-\frac{1}{2}} \left( -e^{\frac{16T_F\tilde{\mu}(0)}{N^2T}} \right). \quad (B3)
\]
Finally, inserting Eq. [10] into Eq. [B3] recovers Eq. [11] given in the text.

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[1] L. Pauling, J. Chem. Phys. 4, 673 (1936)
[2] London, F., J. Phys. Radium 8, 397 (1937)
[3] P. Hund, Annalen der Physik 424, 102 (1938).
[4] M. Büttiker, Y. Imry, and R. Landauer, Physics Letters A 96, 365 (1983)
[5] L. P. Lévy, G. Dolan, J. Dunsmuir, and H. Bouchiat, Phys. Rev. Lett. 64, 2074 (1990)
[6] V. Chandrasekhar, R. A. Webb, M. J. Brady, M. B. Ketchen, W. J. Gallagher, and A. Kleinsasser, Phys. Rev. Lett. 67, 3578 (1991).
[7] D. Mailly, C. Chapelier, and A. Benoit, Phys. Rev. Lett. 70, 2020 (1993)
[8] A. Lorde, R. Johannes Luyken, A. O. Govorov, J. P. Kottas, J. M. García, and P. M. Petroff, Phys. Rev. Lett. 84, 2223 (2000).
[9] H. Kim, W. Lee, S. Park, K. Kyhm, K. Je, R. A. Taylor, G. Nogues, L. S. Dang, and J. D. Song, Nature Publishing Group 7, 1 (2016)
[10] V. D. Pham, K. Kanisawa, and S. Fölsch, Phys. Rev. Lett. 123, 066801 (2019).
[11] A. C. Bleszynski-Jayich, W. E. Shanks, B. Peaudecerf, E. Ginossar, P. von Oppen, L. Glazman, and J. G. E. Harris, Science 326, 272 (2009).
[12] H. Bluhm, N. C. Koshnick, J. A. Bert, M. E. Huber, and K. A. Moler, Phys. Rev. Lett. 102, 136802 (2009).
[13] R. Deblock, R. Bel, B. Reulet, H. Bouchiat, and D. Mailly, Phys. Rev. Lett. 89, 365 (2002).
[14] E. M. Q. Jariwala, P. Mohanty, M. B. Ketchen, and R. A. Webb, Phys. Rev. Lett. 86, 1594 (2001).
[15] A. Fuhrer, S. Lüscher, T. Ihn, T. Heinzel, K. Ensslin, W. Wegscheider, and M. Bichler, Nature 413, 822 (2001).
[16] N. A. J. M. Kleemans, I. M. A. Bominaar-Silkens, V. M. Fomin, V. N. Gladilin, A. G. Taboada, J. M. García, P. Offermans, U. Zeitler, P. C. M. Christiansen, J. C. Maan, J. T. Devreese, and P. M. Koenraad, Phys. Rev. Lett. 99, 146808 (2007).
[17] T. Chakraborty and P. Pietiläinen, Phys. Rev. B 50, 8460 (1994).
[18] S. Ghosh, Advances in Condensed Matter Physics 2013, 1 (2013).
[19] B. Murzaliev, M. Titov, and M. I. Katsnelson, Phys. Rev. B 100, 1 (2019).
[20] S. Viefers, P. Koskinen, P. Singha Deo, and M. Manninen, Physica E: Low-dimensional Systems and Nanostructures 21, 1 (2004).
[21] L. Saminadayar, C. Bauerle, and D. Mailly, in Encyclopedia of Nanoscience and Nanotechnology, edited by H. S. Nalwa (2004) pp. 267–285.
[22] H. Bouchiat and G. Montambaux, J. Phys. France 50, 2695 (1989).
[23] W. E. Shanks, Persistent Currents in Normal Metal Rings, Ph.D. thesis, Yale University (2011).
[24] F. Monticone and A. Ali, Chinese Physics B 23, 047809 (2014).
[25] B. Kanté, Y.-S. Park, K. O'Brien, D. Shuldman, N. D. Lanzillotti-Kimura, Z. Jing Wong, X. Yin, and X. Zhang, Nature Communications 3, 1180 (2012).
[26] K. R. McEnery, M. S. Tame, S. A. Maier, and M. S. Kim, Phys. Rev. A 89, 013822 (2014).
[27] H. Bary-Soroker, O. Entin-Wohlman, and Y. Imry, Phys. Rev. Lett. 101, 321 (2008).
[28] X. Waintal, G. Fleury, K. Kazymyrenko, M. Houzet, P. Schmitteckert, and D. Weinmann, Phys. Rev. Lett. 101, 106804 (2008).
[29] M. G. Viloria, G. Weick, D. Weinmann, and R. A. Jalabert, Phys. Rev. B 98, 1 (2018).
[30] L. Machura, S. Rogozinski, and J. Luczka, Journal of Physics: Condensed Matter 22, 422001 (2010).
[31] S. K. Maiti, Phys. Scr. 73, 519 (2006).
[32] J. F. Weisz, R. Kishore, and F. V. Kusmartsev, Phys. Rev. B 49, 8126 (1994).
[33] H.-F. Cheung, Y. Gefen, E. K. Riedel, and W.-H. Shih, Phys. Rev. B 37, 6650 (1988).
[34] B. L. Altshuler, Y. Gefen, and Y. Imry, Phys. Rev. Lett. 66, 88 (1991).
[35] O. Entin-Wohlman, Y. Gefen, Y. Meir, and Y. Oreg, Phys. Rev. B 45, 11890 (1992).
[36] M.-K. Yip, J.-R. Zheng, and H.-F. Cheung, Phys. Rev. B 53, 1006 (1996).
[37] D. Loss and P. Goldbart, Phys. Rev. B 43 (1991).
[38] E. Hückel, Z. Physik 70, 204 (1931).
[39] M. Rickhaus, M. Jirasek, L. Tejerina, H. Gotfredsen, M. D. Peeks, R. Haver, H.-W. Jiang, T. D. W. Claridge, and H. L. Anderson, Nature Chemistry 12, 236 (2020).
[40] H. Bluhm, J. A. Bert, N. C. Koshnick, M. E. Huber, and K. A. Moler, Phys. Rev. Lett. 103, 026805 (2009).
[41] G. B. Arfken, H. J. Weber, and F. E. Harris, Mathematical Methods For Physicists, seventh ed., A Comprehensive Guide (Academic Press-Elsevier, Waltham, MA, 2013).