Dynes-like superconductivity in thin Al films in parallel magnetic fields

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(Received 24 February 2020; revised 30 June 2020; accepted 2 July 2020; published 17 July 2020)

Recently, the ubiquitously applicable phenomenological Dynes formula for the tunneling density of states of superconductors has been explained by assuming that the electrons scatter on a random magnetic field due to impurities. In the original derivation, only the magnitude of the field was taken as random, but the direction of the field was assumed to be fixed. Here we show that this assumption can be relaxed, and therefore the concept of Dynes superconductivity is not restricted to systems with a preferred direction in spin space. We also address the question whether thin Al films in parallel magnetic fields can be described as Dynes superconductors.

DOI: 10.1103/PhysRevB.102.014508

I. INTRODUCTION

It was realized early on that different types of impurities influence the superconducting state in different ways. In a milestone paper, Anderson gave a nonperturbative argument which shows that neither the thermodynamic properties nor the tunneling density of states of a superconductor will change in presence of time-reversal invariant potential disorder [1]; this type of disorder is therefore called pair-conserving. On the other hand, as shown by Abrikosov and Gorkov within the self-consistent Born approximation, magnetic impurities do lead to such changes [2] and are called pair breaking. In later work, the effect of magnetic impurities was studied extensively, mostly within the self-consistent T-matrix approximation [3] and its refinements [4]. In all these approaches, the self-energy which characterizes the superconductor subject to pair breaking can be found only numerically, except for the trivial cases of vanishing or very strong pair-breaking disorder.

In a recent series of papers, the problem of superconductors with simultaneously present pair-conserving and pair-breaking impurities has been reconsidered within the coherent potential approximation (CPA) [5–8], which is known to provide a very good description of the single-particle properties of disordered metals [9]. Surprisingly, for a Lorentzian distribution of the pair-breaking fields, a simple analytic solution for the Green's function of the superconductor could be found. Making use of this solution, it has been demonstrated that the tunneling density of states is described by the well-known Dynes formula [10]. Long ago, this formula was proposed purely phenomenologically and because of its simplicity it is widely used by the tunneling community until now.

Superconductors described by the Dynes formula have been dubbed Dynes superconductors. The electron spectral functions of Dynes superconductors depend on both the pair-conserving and the pair-breaking scattering rates, and it was demonstrated [6] that they fit the angle-resolved photoemission data in the nodal region of optimally doped cuprates [11]. The optical conductivity of Dynes superconductors also depends on both scattering rates [7] and the theory fits well the recent anomalous data for disordered thin MoN films [12]. Finally, the CPA-based theory of Dynes superconductors has been shown to be thermodynamically consistent [8] and the theory predicts power-law behavior of several observables in the low-temperature limit, even though the gap function is isotropic.

As it stands, the theory of Dynes superconductors models the magnetic impurities by local magnetic fields which have a random magnitude but a fixed orientation. Thanks to this simplification, it was sufficient to make use of the simplest two-component Nambu-Gorkov spinors [5]. However, a fixed orientation of the random fields implies the existence of a special direction in spin space. In view of the experimentally observed broad applicability of the Dynes formula, it is unlikely that this assumption is universally met. The first goal of the present paper is to check whether Dynes superconductivity can also appear in systems where both the magnitude and the orientation of the local magnetic fields are fluctuating. To achieve this goal, we will reformulate the theory in terms of the four-component Nambu-Gorkov spinors. Our second goal is to apply our theory to the high-quality low-temperature data for ultrathin Al films in parallel magnetic fields [13–15]. The specific question that we will be interested in is whether the superconducting state of such films can be described as a Dynes superconductor.

The outline of this paper is as follows. In Sec. II, we describe the model of an impure superconductor which will be studied. In Sec. III we derive, within CPA, the equations which have to be solved. In Sec. IV, we present the solutions to these equations, concentrating on analytically solvable special cases. In Sec. V, we apply our theory to the experimental data from Refs. [13–15]. Finally, in Sec. VI we will conclude.

II. THE MODEL

In the four-component Nambu-Gorkov notation, we introduce the spinor $\alpha_l^\dagger = (c_{l\uparrow}^\dagger, c_{l\downarrow}^\dagger, c_{l\uparrow}, c_{l\downarrow})$, where $c_{l\sigma}^\dagger$ is the creation operator for an electron with spin σ at site l. Our goal will be to find the spatial and temporal Fourier transform of the 4×4 Matsubara Green's function defined by $\hat{G}(l, l', \tau) = -\langle T\alpha_l(-i\tau)\alpha_{l'}^\dagger\rangle$, where τ is imaginary time and T is the time-ordering operator.

For future convenience, let us introduce the following 4×4 matrices:

$$\gamma^0 = \sigma^0\tau^0, \quad \gamma^1 = \sigma^1\tau^1, \quad \gamma^2 = -\sigma^2\tau^2, \quad \gamma^3 = \sigma^0\tau^3,$$

as well as $\vec{\gamma} = \mathrm{diag}(\vec{\sigma}, -\vec{\sigma}^T)$. Here τ^0 and $\vec{\tau} = (\tau^1, \tau^2, \tau^3)$ are the unit matrix and the Pauli matrices in the Nambu (particle-hole) space, while σ^0 and $\vec{\sigma} = (\sigma^1, \sigma^2, \sigma^3)$ are the unit matrix and the Pauli matrices in spin space. The Cartesian components of $\vec{\gamma}$ will be denoted as $\gamma^x, \gamma^y, \gamma^z$.

We consider a single band of electrons characterized by the bare dispersion $\varepsilon_{\mathbf{k}}$, which are Zeeman coupled to an external magnetic field B along the z direction [16]. The 4×4 bare Green's function of the electron with momentum $\mathbf{k}$ and Matsubara frequency ω_n, defined in terms of the spinor $\alpha^\dagger_{\mathbf{k}} = (c^\dagger_{\mathbf{k}\uparrow}, c^\dagger_{\mathbf{k}\downarrow}, c_{-\mathbf{k}\uparrow}, c_{-\mathbf{k}\downarrow})$, reads

$$\hat{G}_0(n\mathbf{k}) = (i\omega_n\gamma^0 - B\gamma^z - \varepsilon_{\mathbf{k}}\gamma^3)^{-1}.$$

The electrons are assumed to interact via a local attractive potential $\mathcal{U}$ which is supposed to generate a spatially uniform mean-field pairing potential Δ, in agreement with scanning tunneling data for the Dynes superconductors [17]. Furthermore, at each lattice site l the electrons are supposed to be subject to two random spatially uncorrelated fields: the pair-conserving potential field U_l and the pair-breaking magnetic field $\vec{V}_l$. Therefore, the total deviation from the bare Hamiltonian is described by $\mathcal{H}' = \frac{1}{2}\sum_l \alpha^\dagger_l \hat{W}_l \alpha_l$, where the local scattering potential $\hat{W}_l$ can be written as

$$\hat{W}_l = \Delta\gamma^2 + U_l\gamma^3 + \vec{V}_l \cdot \vec{\gamma}. \tag{1}$$

The distribution functions for both fluctuating fields are taken mutually uncorrelated and even, for instance, we will assume that $\mathcal{P}(\vec{V}) = \mathcal{P}(-\vec{V})$.

III. CPA EQUATIONS

In CPA, we introduce an averaged electron self-energy $\hat{\Sigma}_n$ which depends only on the Matsubara frequency ω_n, in terms of which the averaged full Green's function reads

$$\hat{G}^{-1}(n\mathbf{k}) = \hat{G}_0^{-1}(n\mathbf{k}) - \hat{\Sigma}_n.$$

Following Janiš [18], in addition to $\hat{\Sigma}_n$, we introduce another independent variable, the averaged local Green's function $\hat{\mathcal{G}}_n$, which also depends only on frequency. In the superconducting state, both $\hat{\Sigma}_n$ and $\hat{\mathcal{G}}_n^{-1}$ depend parametrically on the pairing potential Δ.

Similarly as in Ref. [8], also in the present four-component case, the central object is a functional of the free-energy density $\mathcal{F}[\Delta, \hat{\Sigma}_n(\Delta), \hat{\mathcal{G}}_n^{-1}(\Delta)]$. Here we take the following expression for $\mathcal{F}$:

$$\mathcal{F} = -\frac{T}{2\mathcal{N}}\sum_{n\mathbf{k}} \mathrm{Tr}\ln\left[\hat{G}_0^{-1}(n\mathbf{k}) - \hat{\Sigma}_n\right] + \frac{|\Delta|^2}{\mathcal{U}} + \frac{T}{2}\sum_n\left[\mathrm{Tr}\ln\hat{\mathcal{G}}_n^{-1} - \left\langle\mathrm{Tr}\ln(\hat{\mathcal{G}}_n^{-1} - \hat{W} + \hat{\Sigma}_n)\right\rangle\right], \tag{2}$$

where $\mathcal{N}$ is the number of lattice sites and the angular brackets denote averaging with respect to the fluctuating fields U and $\vec{V}$. The site index in W_l is not written down in Eq. (2), since after averaging $\mathcal{F}$ is independent of l.

The CPA equations can be obtained by taking the functional derivatives of Eq. (2) with respect to $\hat{\Sigma}_n$ and $\hat{\mathcal{G}}_n^{-1}$ and by treating $\hat{\Sigma}_n$ and $\hat{\mathcal{G}}_n^{-1}$ as independent variables. In this way, we obtain

$$\hat{\mathcal{G}}_n = \frac{1}{\mathcal{N}}\sum_{\mathbf{k}}\left[\hat{G}_0^{-1}(n\mathbf{k}) - \hat{\Sigma}_n\right]^{-1},$$
$$\hat{\mathcal{G}}_n = \left\langle\left(\hat{\mathcal{G}}_n^{-1} - \hat{W} + \hat{\Sigma}_n\right)^{-1}\right\rangle. \tag{3}$$

Note that the first of these equations is consistent with the identification of $\hat{\mathcal{G}}_n$ as a local Green's function.

The gap equation can be found by minimizing $\mathcal{F}$ with respect to Δ. To this end, let us note that $\mathcal{F}$ is stationary with respect to $\hat{\Sigma}_n$ and $\hat{\mathcal{G}}_n^{-1}$. Therefore, it suffices to minimize only with respect to the explicit dependence on Δ of the second and last terms (via $\hat{W}$) in Eq. (2). Making use of the CPA Eq. (3), we finally find a BCS-like gap equation:

$$\Delta = -\frac{\mathcal{U}T}{4}\sum_n \mathrm{Tr}(\hat{\mathcal{G}}_n\gamma^2). \tag{4}$$

To summarize, the ultimate goal within CPA is to solve the coupled set of Eqs. (3) and (4).

It turns out that the CPA equations can be solved by the following ansatz for the self-energy:

$$\hat{\Sigma}_n = -i\Theta_n\gamma^0 + \Lambda_n\gamma^z + \Phi_n\gamma^2 + i\Psi_n\gamma^1. \tag{5}$$

Making use of the symmetries of the Green's function described in the Appendix, one can show that $\Theta_n, \Lambda_n, \Phi_n$, and Ψ_n are real functions of the Matsubara frequency. The functions Θ_n and Λ_n describe the spin-independent and the spin-dependent components of the normal self-energy, respectively. They can be shown to satisfy the relations $\Theta_n = -\Theta_{-n}$ and $\Lambda_n = \Lambda_{-n}$. The functions Φ_n and Ψ_n describe the anomalous self-energy of an s-wave superconductor. The component Φ_n corresponds to singlet pairing and it is even in frequency, $\Phi_n = \Phi_{-n}$, whereas Ψ_n corresponds to triplet pairing and it is odd in frequency, $\Psi_n = -\Psi_{-n}$.

For future convenience, let us introduce the spin-resolved renormalized Matsubara frequency $\widetilde{\omega_n^\sigma} = \omega_n + \Theta_n + i\sigma(B + \Lambda_n)$ and the gap function $\widetilde{\Delta_n^\sigma} = \Phi_n + i\sigma\Psi_n$, where $\sigma = \pm 1$, as well as the auxiliary variables

$$x_n^\sigma = \frac{\widetilde{\Delta_n^\sigma}}{\sqrt{\left(\widetilde{\omega_n^\sigma}\right)^2 + \left(\widetilde{\Delta_n^\sigma}\right)^2}}, \quad y_n^\sigma = \frac{\widetilde{\omega_n^\sigma}}{\sqrt{\left(\widetilde{\omega_n^\sigma}\right)^2 + \left(\widetilde{\Delta_n^\sigma}\right)^2}}.$$

Note that for both values of σ we have $(x_n^\sigma)^2 + (y_n^\sigma)^2 = 1$. Also note that $(\widetilde{\omega_n^\sigma})^* = \widetilde{\omega_n^{-\sigma}}$ and $(\widetilde{\Delta_n^\sigma})^* = \widetilde{\Delta_n^{-\sigma}}$, wherefrom it follows that $(x_n^\sigma)^* = x_n^{-\sigma}$ and $(y_n^\sigma)^* = y_n^{-\sigma}$.

In terms of the variables x_n^σ and y_n^σ, the local Green's function can be written as

$$\hat{\mathcal{G}}_n = \frac{\pi N_0}{2}\sum_\sigma\left[-iy_n^\sigma(\gamma^0 + \sigma\gamma^z) - x_n^\sigma(\gamma^2 + \sigma\gamma^1)\right], \tag{6}$$

where N_0 is the normal-state density of states. Similarly, the inverse local Green's function reads

$$\hat{\mathcal{G}}_n^{-1} = \frac{1}{2\pi N_0}\sum_\sigma\left[iy_n^\sigma(\gamma^0 + \sigma\gamma^z) - x_n^\sigma(\gamma^2 + \sigma\gamma^1)\right]. \tag{7}$$

Inserting Eqs. (5)–(7) into the CPA Eqs. (3) and making use of the scattering potential Eq. (1), after averaging over the fluctuating fields, we can compare the coefficients in front of the four γ matrices on both sides of Eqs. (3). In this way, we obtain four self-consistent equations for the four unknown functions Θ_n, Λ_n, Φ_n, and Ψ_n. The explicit form of these equations is too cumbersome to be written down explicitly.

IV. RESULTS

A. Vanishing Zeeman coupling to external field

Let us start by considering the case $B = 0$, when the external magnetic field vanishes. In this case, the ansatz for the self-energy Eq. (5) can be simplified by taking $\Lambda_n = 0$ and $\Psi_n = 0$. As a result, $x_n^\sigma = x_n$ and $y_n^\sigma = y_n$ become independent of the spin projection σ and the components of $\hat{\mathcal{G}}_n$ and $\hat{\mathcal{G}}_n^{-1}$ proportional to γ^z and γ^1 vanish.

After introducing the dimensionless fluctuating fields $\mu = \pi N_0 U$ and $\vec{\lambda} = \pi N_0 \vec{V}$, as well as the dimensionless self-energies $\vartheta_n = \pi N_0 \Theta_n$ and $\delta_n = \pi N_0(\Delta - \Phi_n)$, the self-consistent Eqs. (3) for the unknown functions Θ_n and Φ_n can be written as

$$x_n = (x_n + \delta_n)K_n^-, \quad y_n = (y_n - \vartheta_n)K_n^+, \tag{8}$$

where

$$K_n^\pm = \left\langle \frac{L_n^\pm}{(L_n^-)^2 + 4(y_n - \vartheta_n)^2|\vec{\lambda}|^2 - 4\mu^2|\vec{\lambda}|^2} \right\rangle$$

and the angular brackets denote averaging with respect to the fluctuating fields μ and $\vec{\lambda}$. Moreover, to simplify the formulas, we have introduced

$$L_n^\pm = (x_n + \delta_n)^2 + (y_n - \vartheta_n)^2 + \mu^2 \pm |\vec{\lambda}|^2.$$

Note that the energy scale Δ enters Eqs. (8) only as an external parameter. In a fully self-consistent solution, this scale has to be determined from Eq. (4). Obviously, this task is much simpler when Eqs. (8) can be solved analytically. However, for a general distribution of the fluctuating fields μ and $\vec{\lambda}$, Eqs. (8) for the unknowns Θ_n and Φ_n at a fixed Matsubara frequency ω_n can be solved only numerically. Nevertheless, there do exist special cases when an analytical solution is available and these cases will be treated in what follows:

(i) No pair breaking. If the pair-breaking processes are absent, i.e., in the case $\vec{V} = 0$ which corresponds to $\vec{\lambda} = 0$, one observes readily that $K_n^+ = K_n^- = K_n$. Excluding K_n from Eqs. (8) we find that $\omega_n \widetilde{\Delta}_n = \widetilde{\omega}_n \Delta$. However, if we remember that in terms of the wave-function renormalization Z_n, we can write $\widetilde{\omega}_n = Z_n \omega_n$ and $\widetilde{\Delta}_n = Z_n \Delta_n$, then we find that the energy-dependent gap function Δ_n is not renormalized, $\Delta_n = \Delta$. Consequently, none of the thermodynamic properties of the superconductor is changed with respect to the BCS prediction, including in particular the density of states. This is of course consistent with the Anderson theorem [1].

(ii) Fluctuating magnetic field. Let us assume next that the pair-breaking field $\vec{V}$ as well as the potential field U are nonvanishing. This case is similar to the one studied in previous works on the Dynes superconductors [5,8], but it goes beyond those studies by allowing for fluctuations not only of the magnitude of the internal field $\vec{V}$, but also of its direction, see Fig. 1. In other words, the present case does not assume any anisotropy of the spin space, as has been done in Refs. [5,8]. Nevertheless, remarkably, Dynes superconductivity also arises in this case and the required distribution function is very similar as before: All we have to assume is that the distribution of the *magnitude* of the field $\vec{V}$ is Lorentzian:

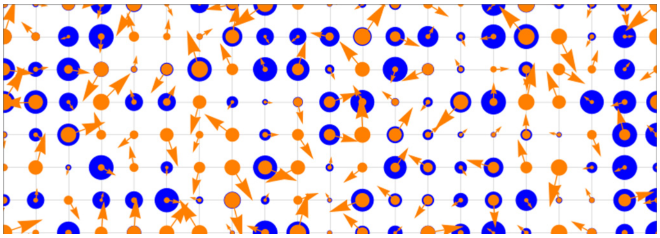

FIG. 1. Schematic view of the disordered lattice. Blue dots: Random pair-conserving field U. Orange arrows: Random pair-breaking magnetic field $\vec{V}$.

$$\mathcal{P}(|\vec{V}|) = \frac{2}{\pi}\frac{\Gamma}{|\vec{V}|^2 + \Gamma^2}.$$

There exist three simple limiting realizations of this distribution: The internal fields may be either completely isotropic, or confined to a plane (easy plane), or lying along a special direction (easy axis). It is the last realization which has been discussed in Refs. [5,8].

Once the magnitude of the field $\vec{V}$ is Lorentzian distributed, the analysis of Eqs. (8) becomes identical to that presented earlier [5] and we recover the known results for Dynes superconductors,

$$\widetilde{\omega}_n = (\omega_n + s_n\Gamma)\left(1 + \frac{\Gamma_s}{\Omega_n}\right), \quad \widetilde{\Delta}_n = \Delta\left(1 + \frac{\Gamma_s}{\Omega_n}\right), \tag{9}$$

where $s_n = \mathrm{sgn}(\omega_n)$ and $\Omega_n = \sqrt{(\omega_n + s_n\Gamma)^2 + \Delta^2}$. Note that the self-energy depends on two scattering rates: the pair-breaking rate Γ which is equal to the width of the Lorentzian distribution $\mathcal{P}(|\vec{V}|)$, and the pair-conserving rate Γ_s which is determined by the distribution function of the potential scatterers $\mathcal{P}_s(U)$ [5]. Only the pair-breaking rate Γ enters the density of states which is given by the Dynes formula:

$$N(\omega) = N_0 \mathrm{Re}\left[\frac{\omega + i\Gamma}{\sqrt{(\omega + i\Gamma)^2 - \Delta^2}}\right]. \tag{10}$$

B. Finite Zeeman coupling to external field

This case is considerably more difficult to solve, since all four self-energies Θ_n, Λ_n, Φ_n, and Ψ_n are nonvanishing. For definiteness, we will assume that the external field lies along the z direction. In what follows, we will consider two special situations:

(i) No pair breaking. Let us assume that the potential scattering U is finite, but the fluctuating magnetic field due to impurities vanishes, $\vec{V} = 0$. In this case, Anderson's theorem applies and the density of states is a sum of BCS-like results shifted by $\pm B$.

(ii) Dynes superconductors. In another and more interesting analytically solvable case, we allow for a finite potential scattering U and, simultaneously, for a finite fluctuating magnetic field V^z due to impurities, polarized in the z direction. If we assume that the distribution function of V^z is Lorentzian with width Γ, then a calculation which closely parallels that in the $B = 0$ case leads to the following results for the self-energy:

$$\widetilde{\omega_n^\sigma} = (\omega_n + i\sigma B + s_n\Gamma)\left(1 + \frac{\Gamma_s}{\Omega_n^\sigma}\right),$$
$$\widetilde{\Delta_n^\sigma} = \Delta\left(1 + \frac{\Gamma_s}{\Omega_n^\sigma}\right), \tag{11}$$

where $\Omega_n^\sigma = \sqrt{(\omega_n + i\sigma B + s_n\Gamma)^2 + \Delta^2}$. Note that these results represent a natural generalization of Eqs. (9) to the case with a finite Zeeman coupling to an external field B. Since both $\widetilde{\omega_n^\sigma}$ and $\widetilde{\Delta_n^\sigma}$ are complex valued, the self-energies Λ_n and Ψ_n are finite. This means that the electrons experience a renormalized magnetic field $\widetilde{B_n} = B + \Lambda_n$ and, at the same time, an odd-frequency triplet pairing field is generated. Expanding to first order in the external field B, we find that $\Lambda_n, \Psi_n \propto \Gamma_s B\Delta$. This means that the nontrivial self-energies Λ_n and Ψ_n are finite only if all three components are present: superconductivity ($\Delta \neq 0$), Zeeman coupling to an external field ($B \neq 0$), and pair-conserving scattering ($\Gamma_s \neq 0$).

In what follows, we will briefly describe the physical properties of the state described by Eqs. (11). Let us start by noting that the density of states is given by the spin-split version of the Dynes formula Eq. (10), $N(\omega) \to \frac{1}{2}\sum_\sigma N(\omega - \sigma B)$. It should be pointed out that this result differs from the classic results obtained in the Born approximation [19–21] in that, in the present case, any finite pair-breaking Γ completely fills the gap, a point we will come to in more detail in the next Section.

The self-consistent Eq. (4) for $\Delta(T, B, \Gamma)$ which is implied by Eqs. (11), takes the simple BCS-like form

$$\Delta = 2g\pi T \sum_{\omega_n>0}^{\Omega_{\max}} \mathrm{Re}\left[\frac{\Delta}{\sqrt{(\omega_n + iB + \Gamma)^2 + \Delta^2}}\right],$$

where $g = N_0\mathcal{U}$ is the dimensionless coupling constant and $\Omega_{\max}$ is the frequency cutoff. Making use of this result and of Eqs. (11) in the free-energy functional Eq. (2), we find the following expression for the condensation energy:

$$\delta\mathcal{F} = -2N_0\pi T \sum_{\omega_n>0}^{\Omega_{\mathrm{Max}}} \mathrm{Re}\left[\frac{[\Omega_n^+ - (\omega_n + iB + \Gamma)]^2}{\Omega_n^+}\right].$$

The knowledge of $\delta\mathcal{F}$ allows us to construct the phase diagrams in the T versus B plane. It is well known that at low temperatures and high fields, the superconductor to normal metal transition is of first order. The equilibrium transition lines $B = B(T)$ determined by solving $\delta\mathcal{F} = 0$ are shown in Fig. 2. As was to be expected, the region where superconductivity is stable shrinks with increasing Γ. Also shown in Fig. 2 are regions of metastability of the normal ($\Delta = 0$) and superconducting ($\Delta \neq 0$) states. Note that these regions shrink with increasing the pair-breaking parameter Γ, and for $\Gamma > 0.355\Delta_{00}$—where Δ_{00} is the gap parameter for $T = 0$ and $B = 0$ of a system without pair breaking—the superconductor to normal metal transition is of second order down to the lowest temperatures.

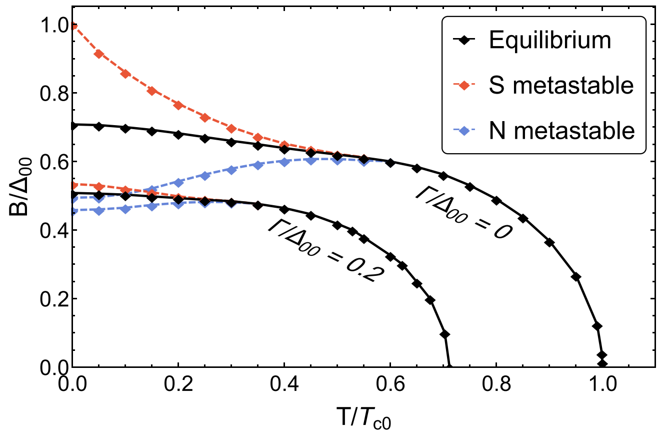

FIG. 2. Phase diagrams in the T versus B plane for a BCS superconductor and a Dynes superconductor with $\Gamma = 0.2\Delta_{00}$. Regions of metastability of the superconducting (S) and normal (N) phase are also shown. T_{c0} is the critical temperature of the BCS superconductor in zero applied field.

V. AL FILMS IN PARALLEL FIELD

In what follows, the results of the previous section will be compared with experimental data for thin superconducting films in parallel magnetic fields, since in this case the orbital coupling to the field can be neglected with respect to the much more important Zeeman coupling to the field. In previous theoretical work, the effect of magnetic impurities as well as that of the spin-orbit scattering on the superconducting properties has been treated in a comprehensive way within the self-consistent Born approximation [19–21]. The main finding of these papers is that, unless the pair-breaking effects are very strong, the superconducting gap remains hard, i.e., the density of states is strictly zero in a finite interval of energies around the Fermi level.

The experimental study of spin splitting in superconductors has a long history. In the classic work on Al films in parallel magnetic fields [21], the authors argue that to fit the tunneling data, it is sufficient to consider the scattering processes in the Born approximation. However, the relatively large value of the thermal smearing of the differential conductance at the experimental temperature $T = 400$ mK of Ref. [21] does not allow us to clearly discriminate between the hard gap predicted by the Born approximation and the soft gap of a Dynes superconductor.

On the other hand, the more recent set of experiments on ultrathin Al films in parallel magnetic fields [13–15], which has been carried out at much lower temperatures, allows us to give a sharp answer to the question whether the superconducting gap is hard or soft. To illustrate this point, in Fig. 3 we show the tunneling conductance of a normal metal-insulator-superconductor (NIS) junction with a superconducting Al electrode in a parallel field $B = 5$ T

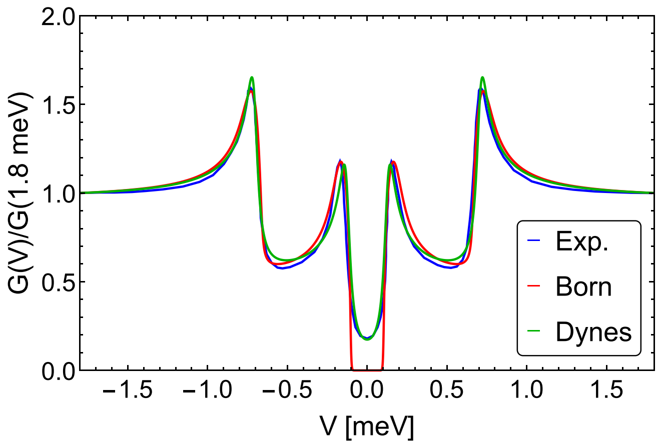

FIG. 3. Tunneling conductance of an NIS junction with a superconducting Al electrode in a parallel field $B = 0.29$ meV, which corresponds to 5 T, at temperature $T = 30$ mK (blue line) [14]. The red curve is a fit which takes the pair-breaking effects into account within the Born approximation. The green curve is a fit using the spin-split Dynes formula. Thermal smearing has been taken into account in both fits. The fitting parameters are shown in the text.

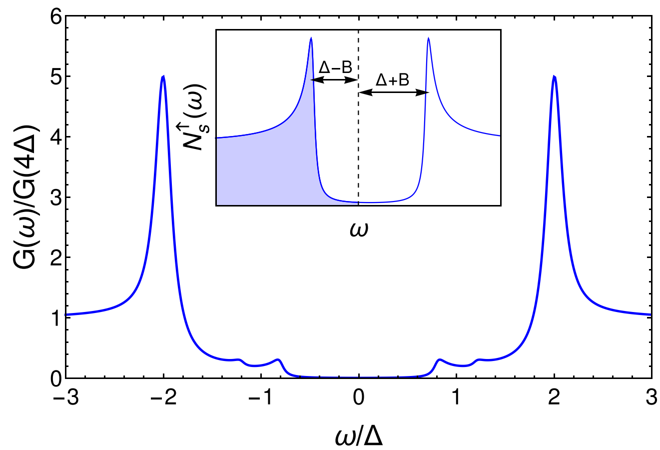

FIG. 4. Low-temperature tunneling conductance of an SIS junction between two identical Dynes superconductors with pair-breaking parameters $\Gamma = 0.05\Delta$ in external magnetic field $B = 0.2\Delta$. Spin-conserving tunneling was assumed. The inset shows processes which generate the subgap peaks at $|\omega| \approx \Delta \pm B$ for spin-up electrons. The shaded area denotes the occupied states below the Fermi level (shown as vertical dashed line). Spin-down electrons generate the same peaks.

at temperature $T = 30$ mK [14]. Two fits of the data are presented in the same figure.

The first fit is based on the standard theory within the Born approximation [19–21]. To achieve a good fit in the superconducting peak region, we have taken $\Delta = 0.43$ meV and two pair-breaking scattering rates: magnetic scattering rate $\Gamma_{\text{mag}} = 0.006$ meV and spin-orbit scattering rate $\Gamma_{\text{s.o.}} = 0.015$ meV. Note that we cannot simultaneously achieve a good fit of both the filled gap and the sharp peaks within this formalism.

The second fit makes use of the spin-split Dynes formula Eq. (10). One observes that with just two fitting parameters, $\Delta = 0.415$ meV and $\Gamma = 0.028$ meV, the fit is of reasonable quality in the whole measured range of energies. To summarize, the analysis of NIS data leads us to conclude that the Al samples can be described as Dynes superconductors.

Unfortunately, the agreement between theory and experiment is spoiled by the data for superconductor-insulator-superconductor (SIS) junctions in applied magnetic fields [15]. In fact, if we assume that Al films in parallel fields are Dynes superconductors, then, taking as usual the tunneling process to be spin conserving, the expected tunneling conductance of the SIS junctions (for identical superconductors on both sides of the junction) is plotted in Fig. 4. As can be seen there, in a Dynes superconductor one should find dominant peaks at $|\omega| \approx 2\Delta$, but also smaller features at $|\omega| \approx \Delta \pm B$. As shown in the inset, these peaks arise from transitions between states in the vicinity of the Fermi level (where the density of states in a Dynes superconductor is finite) and states in the coherence peaks. These processes are completely analogous to the processes which generate the smaller absorption edge at $\omega \approx \Delta$ in the optical conductivity of Dynes superconductors (in zero external field B) [7].

However, although the experimental tunneling conductance of the SIS junctions does exhibit small features within the gap, these are not located at $|\omega| \approx \Delta \pm B$, as expected for a Dynes superconductor, but rather at $|\omega| \approx 2(\Delta - B)$ [15]. Thus, the NIS data [13,14] and the SIS data [15] seem to be mutually inconsistent. One possible way out is to assume that the dominant pair-breaking mechanisms are different in the NIS and SIS samples. This assumption should be, in principle, falsifiable experimentally. However, if this possibility does not happen to be the case, then most likely a new ingredient will have to be added to the analysis presented here.

VI. CONCLUSIONS

In this paper, we have reformulated the theory of the Dynes superconductors within the four-component Nambu-Gorkov notation. Making use of this formalism, we have found the following results:

(i) Recently it has been shown that the ubiquitous Dynes formula, Eq. (10), describes the tunneling density of states in systems with pair breaking modelled by a random internal magnetic field with fluctuating magnitude, but fixed orientation [5]. Here we have generalized this result by showing that the field orientation may also fluctuate and the field distribution may be completely isotropic.

(ii) The theory for Dynes superconductors has been generalized by including the Zeeman coupling to an external magnetic field B. We have found that the combined presence of the finite B field and of pair-conserving scattering causes an admixture of an odd-frequency triplet component to the order parameter. A closed-form formula for the condensation energy in a finite B field has been found and phase diagrams in the B-T plane were constructed; we have shown that the region of first-order transitions between the normal and superconducting states shrinks with increasing the pair-breaking parameter Γ.

(iii) There exists an alternative explanation of the ubiquitous applicability of the Dynes formula Eq. (10) to the tunneling data, according to which the presence of the parameter Γ in Eq. (10) is caused by inelasticity of the tunneling process [22]. To unambiguously discriminate between this explanation and the intrinsic mechanism based on the concept of Dynes superconductivity [5], we propose to measure the tunneling conductance $G(\omega)$ of SIS junctions formed by two identical Dynes superconductors, for instance, break junctions. As should be clear from Fig. 4, in such junctions $G(\omega)$ should exhibit a subgap peak at $|\omega| \approx \Delta$ (in zero applied field). On the other hand, no such peak is to be expected if the mechanism of Ref. [22] is at work.

(iv) We have analyzed the tunneling experiments on ultrathin Al films in parallel magnetic fields, which have been carried out at very low temperatures. We find that the NIS data [13,14] imply that in-gap states should be present in Al, whereas no such states are visible in the SIS data [15]. Further experimental and theoretical work is needed to resolve this discrepancy.

ACKNOWLEDGMENTS

We would like to thank F. Herman for useful discussions. This work was supported by the agency VEGA under Contract No. 1/0640/20.

APPENDIX: SYMMETRIES OF THE GREEN'S FUNCTION

Let us study the Fourier transform of a Green's function with fermionic operators A and B:

$$G(A, B, \omega_n) = -\int_0^\beta d\tau e^{i\omega_n\tau}\langle TA(-i\tau)B\rangle.$$

It is straightforward to prove the identities

$$G(B^\dagger, A^\dagger, \omega_n) = G(A, B, -\omega_n)^*,$$

$$G(B, A, \omega_n) = -G(A, B, -\omega_n).$$

These relations imply the following symmetries of the 4×4 Nambu-Gorkov Green's function:

$$\hat{G}^*_{ij}(\omega_n, \mathbf{k}) = \hat{G}_{ji}(-\omega_n, \mathbf{k}),$$

$$\hat{G}_{ij}(\omega_n, \mathbf{k}) = -\hat{G}_{\bar{j}\bar{i}}(-\omega_n, -\mathbf{k}),$$

where we have introduced the notations $\bar{1} = 3$, $\bar{2} = 4$, $\bar{3} = 1$, and $\bar{4} = 2$.

In addition to these symmetries which are generally present, in an isotropic system we have $\hat{G}(\omega_n, \mathbf{k}) = \hat{G}(\omega_n, |\mathbf{k}|)$. Moreover, if we consider only the Zeeman coupling and neglect the orbital coupling to an external magnetic field B pointing along the z direction, we expect the following symmetries:

$$\hat{G}_{11}(\omega_n, |\mathbf{k}|, B) = \hat{G}_{22}(\omega_n, |\mathbf{k}|, -B),$$

$$\hat{G}_{33}(\omega_n, |\mathbf{k}|, B) = \hat{G}_{44}(\omega_n, |\mathbf{k}|, -B).$$

[1] P. W. Anderson, J. Phys. Chem. Solids **11**, 26 (1959).
[2] A. A. Abrikosov and L. P. Gor'kov, Sov. Phys. JETP **12**, 1243 (1961).
[3] H. Shiba, Prog. Theor. Phys. **40**, 435 (1968).
[4] Ya. V. Fominov and M. A. Skvortsov, Phys. Rev. B **93**, 144511 (2016).
[5] F. Herman and R. Hlubina, Phys. Rev. B **94**, 144508 (2016).
[6] F. Herman and R. Hlubina, Phys. Rev. B **95**, 094514 (2017).
[7] F. Herman and R. Hlubina, Phys. Rev. B **96**, 014509 (2017).
[8] F. Herman and R. Hlubina, Phys. Rev. B **97**, 014517 (2018).
[9] For a review, see R. J. Elliott, J. A. Krumhansl, and P. L. Leath, Rev. Mod. Phys. **46**, 465 (1974).
[10] R. C. Dynes, V. Narayanamurti, and J. P. Garno, Phys. Rev. Lett. **41**, 1509 (1978).
[11] T. Kondo, W. Malaeb, Y. Ishida, T. Sasagawa, H. Sakamoto, Tsunehiro Takeuchi, T. Tohyama, and S. Shin, Nat. Commun. **6**, 7699 (2015).
[12] J. Simmendinger, U. S. Pracht, L. Daschke, T. Proslier, J. A. Klug, M. Dressel, and M. Scheffler, Phys. Rev. B **94**, 064506 (2016).
[13] Wenhao Wu, J. Williams, and P. W. Adams, Phys. Rev. Lett. **77**, 1139 (1996).
[14] V. Yu. Butko, P. W. Adams, and I. L. Aleiner, Phys. Rev. Lett. **82**, 4284 (1999).
[15] P. W. Adams, Phys. Rev. Lett. **92**, 067003 (2004).
[16] Throughout this paper, we measure the magnetic field in energy units, $B = \mu_B B_{\rm phys}$, where μ_B is the Bohr magneton and $B_{\rm phys}$ is the physical magnetic field.
[17] P. Szabó, T. Samuely, V. Haskova, J. Kacmarcik, M. Zemlicka, M. Grajcar, J. G. Rodrigo, and P. Samuely, Phys. Rev. B **93**, 014505 (2016).
[18] V. Janiš, Phys. Rev. B **40**, 11331 (1989).
[19] J. Keller and R. Benda, J. Low Temp. Phys. **2**, 144 (1970).
[20] R. C. Bruno and B. B. Schwartz, Phys. Rev. B **8**, 3161 (1973).
[21] R. Meservey, P. M. Tedrow, and R. C. Bruno, Phys. Rev. B **11**, 4224 (1975).
[22] J. P. Pekola, V. F. Maisi, S. Kafanov, N. Chekurov, A. Kemppinen, Y. A. Pashkin, O. P. Saira, M. Mottonen, and J. S. Tsai, Phys. Rev. Lett. **105**, 026803 (2010).