Letter

Collisionless dynamics of the pairing amplitude in disordered superconductors

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I consider relaxation of the pairing amplitude in a disordered Bardeen-Cooper-Schrieffer (BCS) superconductor in the absence of the two-particle collisions. My main assumption is that nonmagnetic and magnetic disorder scattering rates are much smaller than the value of the superconducting pairing gap Δ_0. I derive a system of nonlinear equations which describe the collisionless relaxation of the pairing amplitude following a quench of the pairing strength. I find that in a superconductor in which scattering on paramagnetic impurities is dominant, the pairing amplitude in a steady state varies periodically with time even for small deviations from equilibrium. It is shown that such a steady state emerges due to scattering on paramagnetic impurities which leads to a decrease in the value of the resonant frequency of the amplitude mode below $2\Delta_0$.

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Introduction. Almost five decades ago, Volkov and Kogan published a theory of collisionless relaxation of the pairing gap in s-wave superconductors [1]. The latter refers to a regime when the relevant time scale for the dynamics far exceeds order parameter relaxation time $\tau_\Delta = \hbar/\Delta_0$, but is much smaller than the relaxation time due to electron-electron collisions $\tau_{\rm ee} \approx \hbar\varepsilon_F/\Delta_0^2$ (ε_F is the Fermi energy). They have considered a model in which the Cooper pairing between the conduction electrons is mediated by their interaction with the acoustic phonons. By focusing on the time scales much longer than inverse value of the energy gap Δ_0 in equilibrium, as well as Debye frequency ω_D, they have derived a system of equations describing the relaxation of an energy gap in clean superconductors. In the linear regime when the deviations from the equilibrium are almost negligible, the time dependence of the pairing amplitude had been found analytically, while the dynamics of the pairing amplitude for stronger deviations from equilibrium remained unknown [1].

A significant progress in the understanding of the collisionless pairing dynamics was made only thirty years after the work of Volkov and Kogan. The interest to this problem has been revived in the context of superfluidity in the atomic condensates [2]. Indeed, in these systems one can induce collisionless dynamics by a sudden change of the magnetic field controlling the optical trap, which inevitably leads to a change of the pairing strength [3]. Soon after that it was realized that the problem of finding the relaxation of the pairing amplitude for an arbitrary deviations from equilibrium admits an exact solution [4–7] (see also Ref. [8] for a comprehensive review). In particular, for strong enough deviations from equilibrium, it was found that the time dependence of the pairing amplitude at long times does not asymptote to a constant value, but remains periodic in time [2,7].

These theoretical developments have triggered the emergence of experiments which aimed to observe the evolution of the energy gap in superconducting films subject to external electromagnetic pulses in terahertz frequency range [9–11]. Theoretical analysis of these experiments, however, typically relies on the results of the early theoretical works [1,8,12]. Importantly, in the context of the methodology used in [1], one usually completely neglects the effects of disorder, which is inevitably present in the superconducting samples and may affect the resulting nature of the steady state when the corresponding time scale due to disorder scattering $\tau_{\rm dis}$ satisfies the condition $\tau_\Delta \ll \tau_{\rm dis} \ll \tau_{\rm ee}$. It is worth mentioning, that the effects of potential disorder has been recently studied in the context of the pump-probe setup [13–17], however, the effects of weak magnetic impurities on the dynamics of the amplitude Higgs mode have never been discussed so far.

In what follows I employ the Keldysh field-theoretical framework to derive a set of nonlinear equations for the dynamics of disordered superconductors in the collisionless regime for the Bardeen-Cooper-Schrieffer (BCS) model of superconductivity [18], including both nonmagnetic and paramagnetic disorder potentials. It is clear that dynamics can only be induced when the initial value of the pairing amplitude is different from the equilibrium one. One way to initiate the dynamics is to assume that the value of the pairing strength has been instantaneously changed [2,4,5], so that by virtue of the self-consistency condition $\Delta(t=0) \neq \Delta_0$. Without loss of generality I will adopt this procedure here as well [12]. Furthermore, from the exact solution of the Volkov-Kogan equations it is known that for small deviations from equilibrium, $\Delta(t)$ approaches a constant value [8] at long times $\tau_\Delta \ll t \ll \tau_{\rm dis}$. This behavior originates from the branch point at $\epsilon = 2\Delta_0$ [1,2,8]. On this time scale the collision integrals, which we evaluate using the exact solution assuming that disorder is weak, should not and, in fact they do not, affect the dynamics. At even longer times $t \sim \tau_{\rm dis} \ll \tau_{\rm ee}$, however, it is not *a priori* clear whether the disorder scattering will produce changes to this steady state and it is precisely the question that I will address in this paper.

In this Letter I demonstrate that in the presence of paramagnetic impurities, the collisionless dynamics remains robust with respect to the dephasing processes, i.e., is dissipationless. Specifically, I find that already in the linear (Volkov-Kogan) regime and for a weak disorder, out-of-equilibrium dynamics of the pairing amplitude is described by a function which

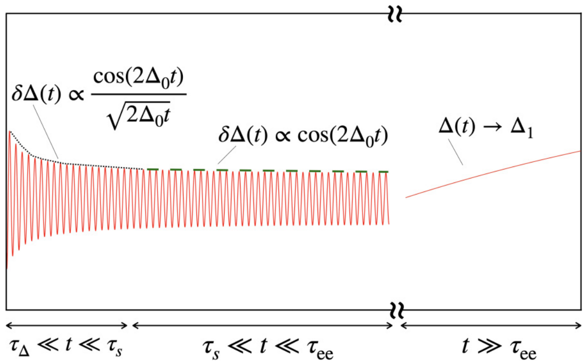

FIG. 1. Time evolution of the pairing amplitude in a conventional superconductor contaminated with a small amount of weak magnetic impurities following an abrupt (but small) change of the interaction strength $|\delta\lambda|/\lambda \ll 1$. The scattering on magnetic impurities is described by a relaxation time τ_s. On a timescale $\tau_\Delta \ll t \ll \tau_s$ the order parameter $\delta\Delta(t) = \Delta(t) - \Delta_0$ oscillates with an amplitude which decays as $1/\sqrt{t}$. However, at longer times $\tau_s \ll t \ll \tau_{\rm ee}$ the order parameter oscillated periodically with time. The amplitude of this oscillations is proportional to $1/(\tau_s\Delta_0)$. On a time scale $t \gg \tau_{\rm ee}$, electron-electron scattering induces relaxation and the pairing amplitude reaches its new equilibrium value.

periodically oscillates with time. This is in stark contrast with the results of the earlier studies where much stronger deviations from equilibrium were required to find such type of a steady state [8]. My result is schematically depicted in Fig. 1.

Model and basic equations. We consider a model with the following Hamiltonian:

$$\hat{H} = \sum_{\alpha\beta}\int d^3\mathbf{r}\overline{\psi}_\alpha(\mathbf{r})[h(-i\vec{\nabla})\delta_{\alpha\beta} + U_{\alpha\beta}(\mathbf{r})]\psi_\beta(\mathbf{r}) - g\int d^3\mathbf{r}\overline{\psi}_\uparrow(\mathbf{r})\overline{\psi}_\downarrow(\mathbf{r})\psi_\downarrow(\mathbf{r})\psi_\uparrow(\mathbf{r}). \tag{1}$$

Here $\psi_\sigma(\mathbf{r})$ is an annihilation operator for a fermion with spin projection $\sigma = \pm 1/2$, $h(-i\vec{\nabla})$ is a kinetic energy operator, g is the coupling constant and the last term accounts for disorder

$$U_{\alpha\beta}(\mathbf{r}) = \sum_j u(\mathbf{r}-\mathbf{r}_j)\delta_{\alpha\beta} + (\vec{S}\cdot\vec{\sigma}_{\alpha\beta})\sum_l J(\mathbf{r}-\mathbf{r}_l). \tag{2}$$

In (2) the summation is performed over the impurity sites and we assume that nonmagnetic and paramagnetic impurities belong to different lattice sites and/or interstitials. The disorder potentials entering into this expression are described by the following correlators $\langle u(\mathbf{r})u(\mathbf{r}')\rangle_{\rm dis} = \delta(\mathbf{r}-\mathbf{r}')/(2\pi\nu_F\tau_u)$ and $S(S+1)\langle J(\mathbf{r})J(\mathbf{r}')\rangle_{\rm dis} = \delta(\mathbf{r}-\mathbf{r}')/(2\pi\nu_F\tau_s)$, where ν_F is the single particle density of states at the Fermi level, S is the spin of a paramagnetic impurity and the averaging is performed over disorder distribution.

Equations of motion for the Green's functions. We consider the fermionic operators on the Keldysh contour and introduce the correlation functions $G_{\alpha\beta}^{(ab)}(1,2) = -i\langle\hat{T}_t\psi_\alpha(1_a)\overline{\psi}_\beta(2_b)\rangle$, where $\psi_1 = \psi_\uparrow$, $\psi_2 = \overline{\psi}_\downarrow$, and $a, b = 1(2)$ refer to the top (bottom) parts of the Keldysh contour. As it directly follows from the definition of $G_{\alpha\beta}^{(ab)}(1,2)$, only three out of four functions (with respect to the Keldysh contour label) are independent: indeed, as it can be directly verified $\hat{G}^{(12)} + \hat{G}^{(21)} = \hat{G}^{(11)} + \hat{G}^{(22)}$. Hence, we consider the retarded, advanced, and Keldysh propagators: $\hat{G}^R(1,2) = \hat{G}^{(11)}(1,2) - \hat{G}^{(12)}(1,2)$, $\hat{G}^A(1,2) = \hat{G}^{(11)}(1,2) - \hat{G}^{(21)}(1,2)$, and $\hat{G}^K(1,2) = \hat{G}^{(11)}(1,2) + \hat{G}^{(22)}(1,2)$. These functions satisfy the following relations $[G_{\alpha\beta}^{R(A)}]^* = -(-1)^{\alpha+\beta}G_{\overline{\alpha}\overline{\beta}}^{R(A)}$, $[G_{\alpha\beta}^K]^* = (-1)^{\alpha+\beta}G_{\overline{\alpha}\overline{\beta}}^K$, which we will use in what follows [1].

Dyson equations. The equations of motion for the functions $G_{\alpha\beta}^{(ab)}(1,2)$ can be derived from the equations of motion for the fermionic operators. As a result one finds that these functions satisfy the Dyson equations:

$$[\hat{G}_0 - \hat{\Sigma}]\circ\hat{G} = \hat{1}, \quad \hat{G}\circ[\hat{G}_0 - \hat{\Sigma}] = \hat{1}, \tag{3}$$

where $\hat{G}_0$ denotes the bare Green's functions for a clean superconductor in the mean-field approximation.

Self-energy parts. Self-energy parts $\hat{\Sigma}$ in Eq. (3) can be obtained by perturbation theory [19]. After performing averaging over disorder and using the correlators for the disorder potential (2) we found

$$\Sigma_{\sigma_1\sigma_2}^{(ij)}(1,2) = \frac{\delta(\mathbf{r}_1-\mathbf{r}_2)}{2\pi\nu_F\tau_s}\left(\hat{\gamma}_{im}^z G_{\sigma_1\sigma_2}^{(mn)}(1,2)\hat{\gamma}_{nj}^z\right) + \frac{\delta(\mathbf{r}_1-\mathbf{r}_2)}{2\pi\nu_F\tau_u}\hat{\sigma}_{\sigma_1\sigma_3}^z\left(\hat{\gamma}_{im}^z G_{\sigma_3\sigma_4}^{(mn)}(1,2)\hat{\gamma}_{nj}^z\right)\hat{\sigma}_{\sigma_4\sigma_2}^z, \tag{4}$$

where the argument of the Green's function should be understood as $(1,2) = (x_1, x_2)$ with $x = (\mathbf{r}, t)$, $\hat{\sigma}^z$ and $\hat{\gamma}^z$ as Pauli matrices which act in Nambu and Keldysh contour spaces correspondingly (see the Supplemental Material [20]). As it follows directly from the definition (4), these functions satisfy the relation $\hat{\Sigma}^{(11)} + \hat{\Sigma}^{(22)} + \hat{\Sigma}^{(12)} + \hat{\Sigma}^{(21)} = 0$. Consequently, we introduce three independent self-energy functions $\hat{\Sigma}^R = \hat{\Sigma}^{(11)} + \hat{\Sigma}^{(12)}$, $\hat{\Sigma}^A = \hat{\Sigma}^{(11)} + \hat{\Sigma}^{(21)}$, and $\hat{\Sigma}^K = \hat{\Sigma}^{(12)} + \hat{\Sigma}^{(21)}$.

Equations of motion for the Keldysh function. Having defined the self-energy part, we are ready to write down the equation of motion for the Keldysh function. This is done in two steps (see the Supplemental Material [20] for details on the derivation). First we obtain the equations of motion with respect to time $t = (t_1 + t_2)/2$:

$$\begin{aligned}&[i\partial_t - h(1) + h^*(2)]G_{11}^K + \Delta(1)\left[G_{12}^K(1,2)\right]^* + G_{12}^K(1,2)\overline{\Delta}(2) = I_{11}^K(1,2),\\ &[i\partial_t - h(1) - h^*(2)]G_{12}^K + G_{11}^K(1,2)\Delta(2) - \Delta(1)\left[G_{11}^K(1,2)\right]^* = I_{12}^K(1,2).\end{aligned} \tag{5}$$

Here we introduced the collision integrals,

$$I_{\alpha\beta}^K(1,2) = \sum_\lambda\left(\Sigma_{\alpha\lambda}^R\circ G_{\lambda\beta}^K - \Sigma_{\alpha\lambda}^K\circ G_{\lambda\beta}^A + G_{\alpha\lambda}^R\circ\Sigma_{\lambda\beta}^K - G_{\alpha\lambda}^K\circ\Sigma_{\lambda\beta}^A\right)(1,2). \tag{6}$$

The second step consists of performing the Wigner transformation with respect to the relative time $\delta t = t_2 - t_1$ and relative position $\delta\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_1$:

$$\check{G}(1,2) = \int\frac{d\varepsilon}{2\pi}\int\frac{d^3\mathbf{p}}{(2\pi)^3}\check{G}(t;\mathbf{p},\varepsilon)e^{i\varepsilon\cdot\delta t - i\mathbf{p}\cdot\delta\mathbf{r}}. \tag{7}$$

We use the same transformation for the collision integrals.

Importantly, in order to compute the Wigner transform of the convolutions $C(1,2)=(A\circ B)(1,2)$ we use $\hat{C}_{\mathbf{p}\varepsilon}(t)=\hat{A}_{\mathbf{p}\varepsilon}(t)e^{\frac{i}{2}(\overleftarrow{\partial}_\varepsilon\overrightarrow{\partial}_t-\overleftarrow{\partial}_t\overrightarrow{\partial}_\varepsilon)}\hat{B}_{\mathbf{p}\varepsilon}(t)\approx\hat{A}_{\mathbf{p}\varepsilon}(t)\hat{B}_{\mathbf{p}\varepsilon}(t)+(i/2)(\partial_\varepsilon\hat{A}_{\mathbf{p}\varepsilon}\partial_t\hat{B}_{\mathbf{p}\varepsilon}-\partial_t\hat{A}_{\mathbf{p}\varepsilon}\partial_\varepsilon\hat{B}_{\mathbf{p}\varepsilon})$. We note that only terms proportional to G_{ab}^K in (6) will have nonvanishing gradient contributions, since, in a steady state that we consider, the retarded and advanced propagators do not depend on t. Higher than linear derivatives of $\Sigma^{R(A)}$ with respect to ε will produce small prefactors upon the integration over $\epsilon_{\mathbf{k}}$ and for this reason their contributions can be ignored. In passing we note that the t-dependent contributions from $\Sigma_{\alpha\beta}^K$ produce an additional small prefactor $\sim t^{-1/2}$ at long times and will also be ignored.

Given the form of the Eqs. (5), it will be convenient to work with the real functions $\vec{S}_{\mathbf{p}}=(S_{\mathbf{p}}^x,S_{\mathbf{p}}^y,S_{\mathbf{p}}^z)$ defined as follows:

$$S_{\mathbf{p}}^z(t)=i\int_{-\infty}^{\infty}\frac{d\varepsilon}{2\pi}G_{11}^K(\mathbf{p},\varepsilon;t),$$
$$S_{\mathbf{p}}^x(t)+iS_{\mathbf{p}}^y(t)=i\int_{-\infty}^{\infty}\frac{d\varepsilon}{2\pi}G_{12}^K(\mathbf{p},\varepsilon;t).\tag{8}$$

These quantities bear a clear analogy with Anderson pseudospins with the only exception that their norm $|\vec{S}_{\mathbf{p}}|$ is not conserved by the evolution (see below).

We now use Eq. (5) to derive the following equations for the components of $\vec{S}_{\mathbf{p}}$:

$$\frac{\partial S_{\mathbf{p}}^x}{\partial t}-2\epsilon_{\mathbf{p}}S_{\mathbf{p}}^y=-\frac{\epsilon_{\mathbf{p}}}{\tau\Delta_{\mathrm{s}}}L_{\mathbf{p}}^y(t),$$
$$\frac{\partial S_{\mathbf{p}}^y}{\partial t}+2\epsilon_{\mathbf{p}}S_{\mathbf{p}}^x+2\Delta(t)S_{\mathbf{p}}^z=-\frac{\epsilon_{\mathbf{p}}}{\tau_{\mathrm{m}}E_{\mathbf{p}}}(\mathcal{W}_{\mathbf{p}}-\cos\theta_{\mathbf{p}})+\frac{2L_{\mathbf{p}}^z(t)}{\tau_s}+\frac{\epsilon_{\mathbf{p}}}{\tau\Delta_{\mathrm{s}}}L_{\mathbf{p}}^x(t),$$
$$\frac{\partial S_{\mathbf{p}}^z}{\partial t}-2\Delta(t)S_{\mathbf{p}}^y=-\frac{2L_{\mathbf{p}}^y(t)}{\tau_s},\tag{9}$$

where $\epsilon_{\mathbf{p}}\in[-\omega_D,\omega_D]$ are the single-particle energy levels, $\tau^{-1}=\tau_s^{-1}+\tau_u^{-1}$, $\tau_{\mathrm{m}}^{-1}=\tau_s^{-1}-\tau_u^{-1}$, $\mathcal{W}_{\mathbf{p}}>0$ is a time-independent function which has a maximum at $\epsilon_{\mathbf{p}}=0$ and decays to zero as $\epsilon_{\mathbf{p}}\to\pm\infty$ (see the Supplemental Material [20]). We have introduced the pairing function $\Delta(t)=-\Delta_{12}(1)$, which in its turn is determined self-consistently by

$$\Delta(t)=\frac{\lambda}{2}\int_{-\omega_D}^{\omega_D}S_{\mathbf{p}}^x(t)d\epsilon_{\mathbf{p}}.\tag{10}$$

Here, $\lambda=g\nu_F>0$ is the dimensionless coupling constant and ω_D is an ultraviolet cutoff, which reflects the retardation effects leading to the onset of superconductivity. In the derivation of the Eq. (9), as well as in (10), we have implicitly assumed that a superconductor is particle-hole symmetric, i.e., we consider a system with constant density of states $\nu(\epsilon)\approx\nu_F$. In Eq. (9) the components of $\vec{L}_{\mathbf{p}}(t)$ describe the solution of the equations of motion in a clean superconductor ($\tau\to\infty$) at long times $\Delta(t\gg\tau_\Delta)=\Delta_{\mathrm{s}}$:

$$L_{\mathbf{p}}^x(t)=\frac{\Delta_{\mathrm{s}}}{E_{\mathbf{p}}}\cos\theta_{\mathbf{p}}+\frac{\epsilon_{\mathbf{p}}}{E_{\mathbf{p}}}\sin\theta_{\mathbf{p}}\cos(2E_{\mathbf{p}}t),$$
$$L_{\mathbf{p}}^y(t)=-\sin\theta_{\mathbf{p}}\sin(2E_{\mathbf{p}}t),$$
$$L_{\mathbf{p}}^z(t)=-\frac{\epsilon_{\mathbf{p}}}{E_{\mathbf{p}}}\cos\theta_{\mathbf{p}}+\frac{\Delta_{\mathrm{s}}}{E_{\mathbf{p}}}\sin\theta_{\mathbf{p}}\cos(2E_{\mathbf{p}}t),\tag{11}$$

where $E_{\mathbf{p}}=(\epsilon_{\mathbf{p}}^2+\Delta_{\mathrm{s}}^2)^{1/2}$ and the components of vector $\vec{L}_{\mathbf{p}}$ satisfy the normalization condition $\vec{L}_{\mathbf{p}}^2=1$. The expressions (11) follow directly from (8) if instead of $G_{\alpha\beta}^K$ we use the expressions for the Keldysh propagators in the steady state (see the Supplemental Material [20]). Given the perturbative nature of the calculation which lead to Eq. (9), I would like to emphasize that the value of the pairing amplitude in the steady state Δ_{s} is taken to be equal to the one for a clean superconductor. We note that the equations of motion (9) do not preserve the norm of $\vec{S}_{\mathbf{p}}$ due to the pair breaking processes induced by the paramagnetic impurities. Finally, the definition of the function $\sin\theta_{\mathbf{p}}$ can be found in the Supplemental Material [20].

Dynamics at long times. We are now ready to investigate the collisionless dynamics following the quench of the pairing strength in a disordered superconductor. We start with the case when only paramagnetic impurities are present in a system, $\tau_u\to\infty$. In equilibrium $S_{\mathbf{p}}^y=0$, while $S_{\mathbf{p}}^x$ and $S_{\mathbf{p}}^z$ can be determined from the equations of motion for $G^{R(A)}$ and are given by

$$S_{\mathbf{p}}^x(0)=\frac{2}{\pi}\int_0^\infty\frac{[R_u-\zeta_s][R_u^3-2\zeta_s]du}{R_u^4[(R_u-\zeta_s)^2+(\epsilon_{\mathbf{p}}/\Delta_0)^2]},$$
$$S_{\mathbf{p}}^z(0)=-\frac{2\epsilon_{\mathbf{p}}}{\pi\Delta_0}\int_0^\infty\frac{[R_u^3-2\zeta_s]du}{R_u^3[(R_u-\zeta_s)^2+(\epsilon_{\mathbf{p}}/\Delta_0)^2]},\tag{12}$$

where we introduced function $R_u=\sqrt{1+u^2}$ and dimensionless rate $\zeta_s=1/2\tau_s\Delta_0$ for brevity. Furthermore, using Eq. (9) it can be directly shown that in equilibrium the pseudospin components must satisfy $\epsilon_{\mathbf{p}}S_{\mathbf{p}}^x+\Delta_0S_{\mathbf{p}}^z=-w_{\mathbf{p}}\epsilon_{\mathbf{p}}/\tau_s(\epsilon_{\mathbf{p}}^2+\Delta_0^2)^{1/2}$, where $w_{\mathbf{p}}$ is a known function of $\epsilon_{\mathbf{p}}$ and Δ_0 is the energy gap in equilibrium computed using the self-consistent Born approximation (see the Supplemental Material for details [20]).

The results of the numerical solution of Eq. (9), following small quenches of the pairing strength, are shown in Fig. 2. We immediately observe that the steady state with the oscillating $\Delta(t)$ emerges at times $t\sim\tau_s$. We also notice that the amplitude of the oscillations is proportional to ζ. These results appear to be quite generic with respect to the magnitude and sign of the quench (i.e., when the pairing strength is slightly decreased) as well as initial conditions. We note that a similar result has been recently reported, where the nondissipative Higgs mode appears due to the presence of long-range interactions in a superconductor coupled to a strongly driven cavity [21]. Note also, that the amplitude of the oscillations is parametrically bigger for smaller values of the pairing gap in equilibrium, Fig. 3. The full calculation of the steady state diagram for quenches of an arbitrary strength, as well as strong disorder, we leave for the future studies.

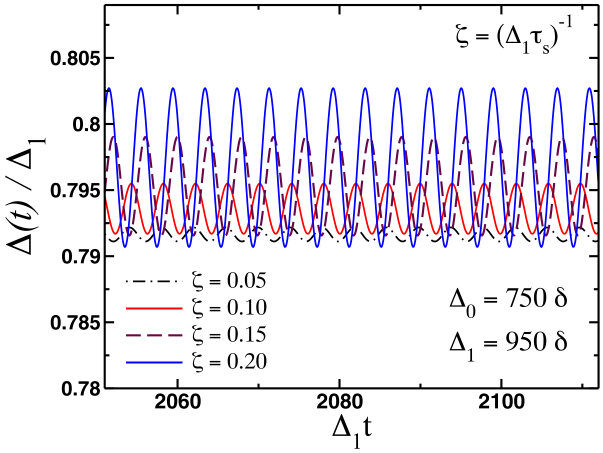

FIG. 2. Results of the numerical solution of the Eqs. (9) for the pairing amplitude $\Delta(t)$ at long times for various values of the dimenionless parameter $\zeta = 1/\tau_s\Delta_1$ which quantifies the strength of paramagnetic disorder. These results have been obtained for the system of $N = 15\,024$ equally spaced energy levels with the level spacing δ. The dynamics was initiated by a sudden change of the dimensionless coupling constant from a value corresponding to the ground state with the pairing gap Δ_0 to a new value corresponding to a ground state with the pairing gap Δ_1.

Discussion. The appearance of the periodically oscillating solution can be understood as follows. Qualitatively, one may interpret this result an a way similar to the interpretation given in Ref. [22]: scattering induced by the paramagnetic impurities pushes the frequency of the amplitude mode inside the energy gap, so that the dephasing is fully suppressed and the mode becomes undamped. Indeed, simple calculation shows that for the frequency of the Higgs mode in this case ($\tau_u \to \infty$) we find

$$\frac{\omega_{\rm Higgs}}{2\Delta_0} = \left[1 - \left(\frac{1}{\tau_s\Delta_0}\right)^2\right]^{1/2}. \tag{13}$$

The time dependence of the pairing amplitude will now be given by

$$\delta\Delta(t) \approx \zeta e^{i\omega_{\rm Higgs}t} + \int_{-\infty}^{\infty}\frac{d\omega}{2\pi}A(\omega)e^{i\omega t}. \tag{14}$$

The last term in this expression decays as $t^{-1/2}$ at long times and so only the first terms contribute.

Equation (13) shows that contrary to our expectations, in a superconductor with a pairing amplitude Δ_0, it takes less than $2\Delta_0$ amount of energy to excite a Cooper pair. Let us also recall that in a superconductor contaminated with paramagnetic impurities, there is another energy scale $\Delta_{\rm th}$ which represents the threshold for the single-particle excitations, i.e., the energy when the single particle density of states becomes nonzero for the first time [23] and I found that $\omega_{\rm Higgs} > 2\Delta_{\rm th}$ (see the Supplemental Material [20]). Therefore, Eq. (13) introduces a completely new energy scale which describes the softening of the "mass" of the Higgs mode. It seems that the physical processes which lead to the appearance of this energy scale are the same as the ones responsible for the appearance of $\Delta_{\rm th}$.

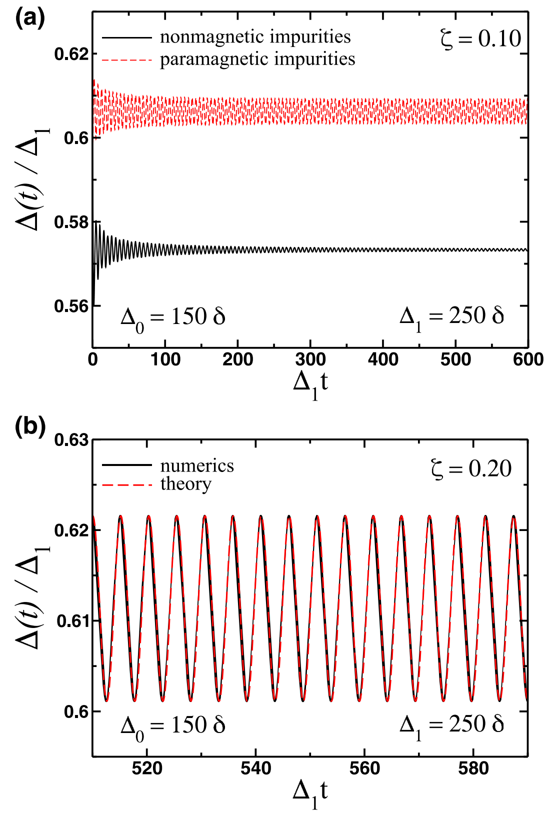

FIG. 3. Panel (a): Solution of the equations of motion for the two separate cases of (i) purely paramagnetic ($\tau_u \to \infty$) and (ii) purely nonmagnetic ($\tau_s \to \infty$) disorder. In the former case $\zeta = 1/\tau_s\Delta_1$, the pairing amplitude periodically oscillates with time, while in the latter case $\zeta = 1/\tau_u\Delta_1$ the amplitude of the oscillations decays as $t^{-1/2}$. Panel (b): Comparison between the results of the numerical solution for $\Delta(t)$ (paramagnetic disorder only) and the phenomenological expression $\Delta(t) = \Delta_0 - \zeta[A + B\cos(2\Delta_0 t + \varphi)]$. We used the following values of the parameters: $A \approx 0.39$, $B \approx 0.01$, and $\varphi \approx \pi/4$. We would like remind the reader that these results hold on time scales shorter than $\tau_{\rm ee}$.

In contrast, for a system in which only potential impurities are present ($\tau_s \to \infty$), for the frequency of the Higgs mode we found

$$\frac{\omega_{\rm Higgs}}{2\Delta_0} = \left[1 + \left(\frac{1}{\tau_u\Delta_0}\right)^2\right]^{1/2}, \tag{15}$$

which lies above the edge of the single particle continuum. This means that in this case the Higgs mode will become dissipative due to dephasing processes and, as a result, pairing amplitude asymptotes to a constant, $\tilde{\Delta}_s$, at long times as shown in Fig. 3(a) (see also discussion in [17]).

For the case when both nonmagnetic and magnetic disorder is present, we find that the frequency of the amplitude mode falls below $2\Delta_0$ when $\tau_{\rm m} > 0$, and is above $2\Delta_0$ when $\tau_{\rm m} < 0$. This latter conclusion, which is based on the perturbative calculation, implies that the Higgs mode will become overdamped in either ballistic or dirty limits when $\tau_u \ll \tau_s$. As it turns out, in general this is not the case and it can be shown that even in diffusive superconductors the frequency of the resonant Higgs mode is less than $2\Delta_0$.

Comparison between these two behaviors, which are governed by Eqs. (13) and (15), is illustrated in Fig. 3. In passing we note that the Yu-Shiba-Rusinov bound states should not affect our results for the dynamics since the wave functions describing these states remain orthogonal to the wave functions corresponding to the continuous spectrum of the single-particle excitations. The same argument also applies to the existence of the tail states in the single particle density of states which are induced by the scattering on nonmagnetic impurities [24]. However, these effects may produce a shift in the value of $\omega_{\rm Higgs}$ as well as the broadening of the Higgs resonance [25]. The detailed study of these effects, as well as the question of whether these states affect the dynamics on the level of the collision integrals, will be addressed separately.

Finally, I note that apart from the fact that the physics I just described can be observed at time scales $t \ll \tau_{\rm ee}$, Fig. 1, it is also important to keep in mind that the steady state with the periodically oscillating pairing amplitude is intrinsically unstable toward developing spacial inhomogeneities [26] when the characteristic size of a sample L is much larger than the coherence length $\xi = v_F/\Delta_0$. Thus, for the pairing amplitude to remain spatially inhomogeneous in a steady state with periodic oscillations, the value of the superconducting order parameter in equilibrium should be sufficiently small, so that the condition $\xi \gtrsim L$ is fulfilled.

Conclusions. In this work I have considered a problem of collisionless relaxation in a superconductor contaminated with nonmagnetic and paramagnetic impurities. I found that in the case when scattering on paramagnetic impurities is dominant, for even small deviations from equilibrium the corresponding steady state is described by the periodically oscillating pairing amplitude, which is an unambiguous manifestation of the amplitude (Higgs) mode in a superconductor.

It is well known that in a superconductor contaminated with paramagnetic impurities with the order parameter Δ_0, it costs $\Delta_{\rm th} < \Delta_0$ to create a single particle excitation [23]. The second main result of this Letter is that in this case it also costs $\omega_{\rm Higgs} < 2\Delta_0$ amount of energy to excite a Cooper pair.

My results provide an avenue for detecting the dynamics of the amplitude Higgs mode in s-wave superconductors. Specifically, recent experimental studies of a superconductor NbN have convincingly demonstrated that in the pump-probe experimental setup, the intensity of the terahertz signal is peaked at $\omega_{\rm peak} = 2\Delta_0$ [25]. Based on the results of this work, I predict that by introducing a small to moderate amount of paramagnetic impurities into NbN film and subjecting it to the electromagnetic pulse in the terahertz range of frequencies, one should observe the shift of the peak in the intensity of the signal from the expected value of $2\Delta_0$ to a smaller value $\omega_{\rm peak} < 2\Delta_0$. If several samples which differ by the amount of magnetic impurities, one should observe the decrease in the ratio $\omega_{\rm peak}/2\Delta_0$ with an increase in the impurity concentrations. By combining this result with the results of the measurements which directly probe the single-particle density of states, one then can compare the value of $\omega_{\rm peak}$ to $2\Delta_{\rm th}$ and verify that $\omega_{\rm peak} > 2\Delta_{\rm th}$. In cases when there is an agreement between the experimental results and my theoretical predictions, it opens up a possibility for direct observation of the dynamics of the amplitude Higgs mode since, as it has been demonstrated in this work, the shift in the frequency of the Higgs mode means that its dynamics becomes undamped on a time scale $t \ll \tau_{\rm ee}$. Experimentally it may be challenging to introduce paramagnetic impurities in a film such that, for example, $\tau_{\rm ee}(T) \sim 10\tau_s \sim 100\tau_\Delta$. Even though making $\tau_{\rm ee}(T)$ large should not be challenging since it is proportional to $\hbar/T$ due to the Al'tshuler-Aronov effect, I am aware of the fact that the requirements for observing the pairing amplitude dynamics are much more stringent due to the difficulties associated with controlling the value of the ratio of the relaxation times τ_s/τ_Δ and τ_u/τ_Δ experimentally.

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Correction: Support information in the last sentence in the Acknowledgment section has been fixed.