

## Coherent Excited States in Superconductors due to a Microwave Field

A. V. Semenov,<sup>1,2,\*</sup> I. A. Devyatov,<sup>3,2,†</sup> P. J. de Visser,<sup>4,5</sup> and T. M. Klapwijk<sup>4,1,‡</sup>

<sup>1</sup>Moscow State Pedagogical University, 1 Malaya Pirogovskaya Street, Moscow 119992, Russia

<sup>2</sup>Moscow Institute of Physics and Technology, Dolgoprudny, Moscow 141700, Russia

<sup>3</sup>Lomonosov Moscow State University, Skobeltsyn Institute of Nuclear Physics, 1(2), Leninskie gory, GSP-1, Moscow 119991, Russia

<sup>4</sup>Kavli Institute of NanoScience, Faculty of Applied Sciences, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands

<sup>5</sup>SRON Netherlands Institute for Space Research, Sorbonnelaan 2, 3584 CA Utrecht, The Netherlands

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We describe theoretically the depairing effect of a microwave field on diffusive *s*-wave superconductors. The ground state of the superconductor is altered qualitatively in analogy to the depairing due to a dc current. In contrast to dc depairing, the density of states acquires, for microwaves with frequency  $\omega_0$ , steps at multiples of the photon energy  $\Delta \pm n\hbar\omega_0$  and shows an exponential-like tail in the subgap regime. We show that this ac depairing explains the measured frequency shift of a superconducting resonator with microwave power at low temperatures.

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How is the superconducting state modified by a current, i.e., when the condensate is moving? The answer to this question is well known for the case of a dc current flowing in a superconducting wire. For a dc current, the Cooper pairs gain a finite momentum which leads to the suppression of the superconducting properties of the wire [1,2]. The modulus of the superconducting order parameter  $\Delta$  is reduced and the sharp BCS singularity near the gap is smeared. This depairing effect of a current or of a magnetic field was studied theoretically soon after the creation of the microscopic theory of superconductivity [3]. The moving superconducting condensate has been called a coherent excited state generated by the momentum displacement operator  $\rho_q = \sum_n \exp(i\mathbf{q} \cdot \mathbf{r}_n)$  by Anderson [4] as part of the explanation of the Meissner effect from the original form of the BCS theory. The momentum displacement operator, when applied to the BCS ground state, creates excited pairs of electrons  $\mathbf{k}_1, \mathbf{k}_2$  with the momentum pairing  $\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{q}$  instead of zero [1,4–6]. This momentum displacement  $q = |\mathbf{q}|$  corresponds to a superfluid drift velocity  $v_s = \hbar q/m$ , where  $m$  is the electron mass. In the Green's function technique, it is possible to introduce the superfluid velocity in a gauge-invariant way,  $v_s \propto [\nabla\varphi - (2e/\hbar)\mathbf{A}]$ , where  $\varphi$  is the phase of the superconductor,  $e$  the electron charge, and  $\mathbf{A}$  the vector potential of the electromagnetic field. The equivalence of depairing due to an electric current and due to a magnetic field is well established, both theoretically [7] and experimentally [2], using thin and narrow superconducting wires with a uniform current density. The theory of depairing by a dc current was reformulated, using the Usadel equations [8], for diffusive films with an elastic scattering length much smaller than the BCS coherence length [9]. The results of this theory [9] were confirmed experimentally by Romijn *et al.* [10] and by Anthore *et al.* [2].

However, a general theory for *depairing by a microwave field*, a time-dependent vector potential  $\mathbf{A}$ , has not been formulated. In current experimental research, there are many cases in which a superconductor is used at very low temperatures,  $T/T_c \ll 1$ , where the density of quasiparticles is very low and the response of the superconductor is dominated by the response of the superfluid. At higher temperatures, it is well known that microwave radiation can be absorbed by quasiparticles, leading to a nonequilibrium distribution over the energies [11]. At very low temperatures, there are hardly any quasiparticles, and with  $\hbar\omega_0 \ll 2\Delta$  there is not enough energy per photon to break Cooper pairs. In practice it has been demonstrated that, in this regime, the microwave power has a significant influence on the superconducting state. For example, de Visser and co-workers [12,13] have measured for aluminium a shift in kinetic inductance and an increase in the density of quasiparticles as a function of microwave power at temperatures of 60 mK. This dependence can be parametrized by labeling it “absorbed power,” but the fundamental question is how the superconducting state responds to a time-dependent electromagnetic field. In previous works only the limiting case of high temperatures (close to the critical temperature  $T_c$ ) [14] or relatively low frequency compared to the temperature ( $\hbar\omega_0 \ll k_B T$ ) [15] were studied. In these cases the coherent properties of a superconductor (e.g., the density of states) change analogous to magnetic impurities and a static magnetic field. However, in general it is to be expected that an oscillating vector potential would lead to coherent excited pairs with an oscillating center of mass motion, with a more substantial modification of the density of states. The change of the energy spectrum of electrons, dressed by an electromagnetic field, is, in principle, analogous to the dynamic Stark effect in atom physics [16] (and similarly for a two-dimensional electron gas

[17]). Such dressed states have recently been put forward in the analysis of the microwave response of superconducting atomic point contacts [18,19]. The relevance of the problem for a plain superconductor is apparent in the case of superconducting parametric amplifiers [20] and in the nonlinearity of kinetic inductance devices with microwave readout [13,21,22]. Additionally, the effects of direct depairing at these frequencies is also of interest for quantum optics on superconducting artificial quantum systems [23,24]. In summary, there is an urgent need for a microscopic theory which correctly describes the depairing of a conventional superconductor by microwave radiation.

We focus on the influence of high-frequency radiation on a narrow and thin dirty  $s$ -wave superconducting strip using the well-established theory of nonequilibrium superconductivity [25,26]. Diffusive motion implies that the direction of momentum on the period of the microwave field is randomized. The thickness and width are assumed to be much less than the relevant penetration depth  $\lambda$ , allowing for a uniform current over the cross section. The strip is assumed to be long enough in order to ignore the influence of the ends of the strip. We consider the case of a relatively high frequency  $\omega_0$  of the microwave radiation:

$$\alpha \ll \hbar\omega_0 \ll 2\Delta, \quad (1)$$

where the parameter  $\alpha$  is the same as in the Eliashberg theory [11]:  $\alpha = e^2 D E_0^2 / \hbar\omega_0^2$ , with  $D$  being the diffusion coefficient and  $E_0$  the amplitude of the microwave field. The left inequality of Eq. (1) is known as the condition for the quantum regime of absorption [11,27–29], and by analogy we call this regime of depairing the “quantum regime of depairing.” The right inequality of Eq. (1) points out that it is not possible to excite quasiparticles directly. Hence, the depairing is only associated with the reconstruction of the ground state of the superconductor in the presence of a microwave field.

We start from the usual mean-field Hamiltonian of a superconductor in the presence of electromagnetic radiation, modeled by a vector potential  $\mathbf{A}$ , introduced in the gauge-invariant form in the kinetic part of this Hamiltonian:  $H_k = -(\hbar^2/2m)\partial^2$ ,  $\partial = \partial/\partial\mathbf{r} - ie\mathbf{A}\check{\tau}_z$ , where  $\check{\tau}_z$  is the third Pauli matrix, operated in Keldysh-Nambu spaces [25,26]. We use the Keldysh-Usadel approach [25,26,30] to determine the response of the superconductor. For details, we refer to the Supplemental Material [31]. Under the condition of Eq. (1), the spatially homogeneous dirty  $s$ -wave superconductor can be described by a closed set of equations containing only the stationary components of the Green’s functions and the order parameter. The stationary components of the retarded Green’s functions satisfy, in the presence of a dc current and in the presence of monochromatic radiation, the equation

$$-iEF_0^R - i\Delta G_0^R + \Pi = 0. \quad (2)$$

In Eq. (2),  $G_0^R = G_0^R(E)$  and  $F_0^R = F_0^R(E)$  are energy ( $E$ ) dependent stationary normal and anomalous components of the superconducting matrix Green’s function  $\hat{G}^R(E)$  in Nambu space. In the case of depairing by a dc current [2], the depairing term  $\Pi$  in Eq. (2) has the form  $\Pi = \Gamma G_0^R F_0^R$ , where  $\Gamma$  is the depairing energy determined by the current density. In the present case of monochromatic radiation, modeled by a vector potential  $A(t) = A \cos(\omega_0 t)$ , the depairing term  $\Pi$  is given by

$$\Pi = \alpha \{ F_0^R (G_{0+}^R + G_{0-}^R) + G_0^R (F_{0+}^R + F_{0-}^R) \}. \quad (3)$$

Equation (3) constitutes the formal difference of depairing by an rf current from the case of depairing by a dc current.  $G_{0\pm}^R = G_0^R(E \pm \hbar\omega_0)$  and  $F_{0\pm}^R = F_0^R(E \pm \hbar\omega_0)$  are both shifted in energy by  $\hbar\omega_0$  with respect to the normal and anomalous component of the Green’s function.  $G_0^R$  and  $F_0^R$  are linked via the normalization condition

$$(G_0^R)^2 - (F_0^R)^2 = 1. \quad (4)$$

The order parameter  $\Delta$ , also stationary, satisfies the self-consistency equation of the usual form

$$\Delta = -\lambda_{ep} \int_0^{\hbar\omega_D} dE (1 - 2f_0) \text{Re} F_0^R, \quad (5)$$

and is thus field dependent, where  $f_0 = f_0(E)$  is the stationary component of the distribution function,  $\omega_D$  the Debye frequency, and  $\lambda_{ep}$  the electron-phonon coupling constant. In the case of excitations by microwaves, the distribution function is symmetric in energy,  $f(E) = f(-E)$ ; i.e., only the longitudinal part  $f_L$  of the distribution function arises:  $\text{sgn}(E)[1 - 2f(E)] = f_L(E)$  [25,26].

This set of equations [Eqs. (2)–(5)] has been solved numerically, the procedure is described in the Supplemental Material [31]. Figure 1 shows the results for the density of states,  $N(E) = N_0 \text{Re} G_0^R(E)$ , for different microwave intensities and frequencies (here,  $N_0$  is the density of states in the normal metal per spin). The black dash-dotted line in Fig. 1(a) corresponds to the pure BCS case without any depairing,  $\alpha = \Gamma = 0$ . The black solid line corresponds to depairing by a dc current with the value of the depairing parameter  $\Gamma = 0.014\Delta_0$ .  $\Delta_0$  is the modulus of the order parameter at zero temperature without depairing ( $\alpha = 0$ ). The red curve corresponds to depairing by microwaves for  $\alpha = 0.0017\Delta_0$ , with the photon energies clearly visible. To facilitate a quantitative estimate, we express the ratio  $\Gamma/\Delta_0$  as the ratio of the dc supercurrent  $j$  to the critical pair-breaking current  $j_c$  with  $\Gamma/\Delta_0 \cong 0.11(j/j_c)^2$ . Similarly, for  $\alpha$ ,  $\alpha/\Delta_0 \cong 0.014(j_0/j_c)^2$ , with  $j_0$  being the amplitude of the rf supercurrent. The dc current and rf current curves in Fig. 1(a) correspond to the same value of  $j$ ,  $j_0 = 0.25j_c$ . Figure 1(b) presents a comparison between densities of

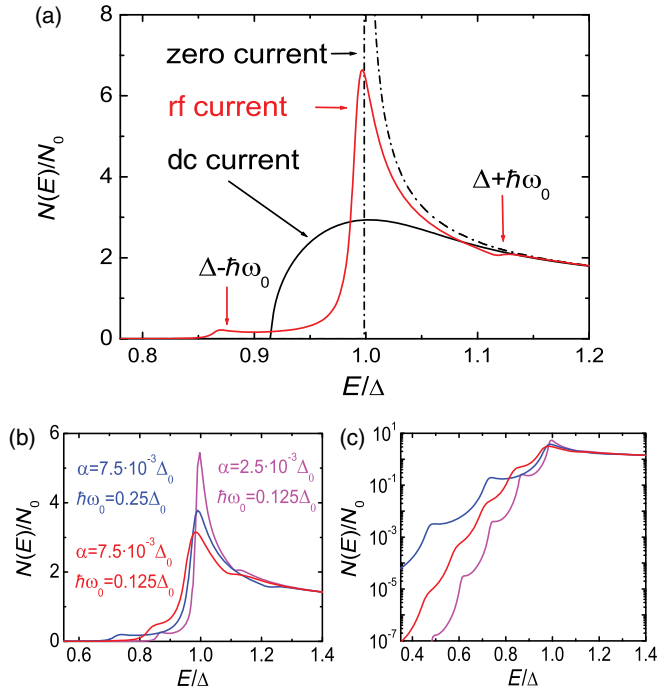


FIG. 1. The normalized density of states of a diffuse superconducting strip  $N(E)/N_0 = \text{Re}G_0^R(E)$ , subject to a monochromatic microwave signal. The black dotted line in (a) corresponds to the pure BCS case without any depairing  $\alpha = \Gamma = 0$ . The black solid line corresponds to depairing by a dc current with the value of the depairing parameter  $\Gamma = 0.014\Delta_0$ . The red curve in (a) corresponds to rf depairing with the value of the parameter  $\alpha = 0.0017\Delta_0$ . The red, blue, and magenta curves in (b) and (c) correspond to rf depairing with different values of  $\alpha$  and for different frequencies: the red curve corresponds to  $\alpha = 0.0075\Delta_0, \hbar\omega_0 = 0.125\Delta_0$ , the blue curve corresponds to  $\alpha = 0.0075\Delta_0, \hbar\omega_0 = 0.25\Delta_0$ , and the magenta curve corresponds to  $\alpha = 0.0025\Delta_0, \hbar\omega_0 = 0.125\Delta_0$ . (c) shows on a logarithmic scale the presence of the low-lying states, which are absent for a zero or dc current, and the steps at multiples of  $\hbar\omega_0$ .

states for different values of the depairing parameter  $\alpha$  and different frequencies, as indicated in the figure.

As is evident from Fig. 1, the depairing due to the rf signal (the red, blue, and magenta curves) is very different from the depairing due to a dc current (the black curve), although both smear the BCS singularity at  $E = \Delta_0$ . One striking difference is that the density of states, which corresponds to dc depairing, goes to zero at some value of energy, whereas the densities of states which correspond to rf depairing have exponential-like tails at small energies [shown in Fig. 1(c)]. In addition, with rf depairing there are irregularities in the density of states at equal distance from each other, corresponding to the photon energy  $\hbar\omega_0$  (Fig. 1). These photon steps are reminiscent of the photon-assisted tunneling steps in the quasiparticle current of tunnel junctions irradiated by a microwave signal [28,29]. In the present case for a plain superconducting film and frequencies corresponding to the right inequality

in Eq. (1), the photon structures in the density of states can be interpreted as a manifestation of dressed states of diffusely scattering electrons, which inevitably must exist in a superconductor in a microwave field. This would naturally arise in the displacement operator  $\rho_q$  by replacing a time-independent  $q$  by  $q_0 \cos \omega t$ . Here, this is accomplished for a diffusive superconductor leading to Eqs. (2) and (3).

Qualitatively, the result reflects that the dressed electron states become superpositions of states with energies shifted by multiples of  $\hbar\omega_0$ . The transition probability of a diffusely moving electron from the state with energy  $E$  to the state with energy  $E \pm \hbar\omega_0$  per unit time is of the order  $\alpha/\hbar$  [11]; thus, during the oscillation period of the field,  $2\pi/\omega_0$ , the components with energy shifted to  $\pm\hbar\omega_0$  acquire a weight of the order  $\alpha/\hbar\omega_0$ . Transitions to  $\pm n\hbar\omega_0$  are processes of the  $n$ th order and the corresponding weights are of the order  $(\alpha/\hbar\omega_0)^n$ . From this consideration, it follows that the BCS peak in the density of states resurfaces in the series of peaks at multiples of the photon energy  $\Delta \pm n\hbar\omega_0$  (photon points), of which the amplitudes decrease exponentially with the growth of  $n$ .

Apart from its fundamental interest, this theoretical analysis is relevant for experiments on microwave kinetic inductance detectors and parametric amplifiers based on superconducting films at millikelvin temperatures, where the electrodynamic response is dominated by the superconducting condensate. Typically, the quantity which is measured is the impedance or the complex conductivity. The conductivity  $\sigma(\omega)$  at frequency  $\omega$  can be derived from the general formula for the current density in a diffusive superconductor in a straightforward way [31]:

$$\sigma(\omega) = \frac{\sigma_N}{4\omega} \int dE \{ [(G_{0-}^R)^* \text{Re}G_0 + (F_{0-}^R)^* \text{Re}F_0] f_{L0} + [G_0^R \text{Re}G_{0-}^R + F_0^R \text{Re}F_{0-}^R] f_{L0-} \}, \quad (6)$$

with  $\sigma_N = 2e^2 N_0 D$  being the conductivity of the wire in the normal state and  $f_{L0-} = f_{L0}(E - \hbar\omega_0)$ . In deriving Eq. (6), we ignore higher order terms leading to a modulation of the conductivity at the doubled signal frequency. Figures 2(a) and 2(b) show the real part of conductivity  $\text{Re}\sigma(\omega)$ , calculated from Eq. (6) with the same values of the depairing parameters  $\alpha, \Gamma$  and signal frequencies  $\omega_0$  as used in Figs. 1(b) and 1(c), for the densities of states. In Figs. 2(a) and 2(b) one observes, qualitatively, the same behavior in the real part of the conductivity  $\text{Re}\sigma(\omega)$  as for the densities of states: the microwave radiation leads to the appearance of anomalies at ‘‘photon points’’ and an exponential-like tail at small values of  $\omega$ .

In a recent experiment [12], a superconducting resonator of aluminium was measured to determine the quality factor and the resonant frequency as a function of applied microwave power. In the interpretation, the absorption by quasiparticles was included through the well-known

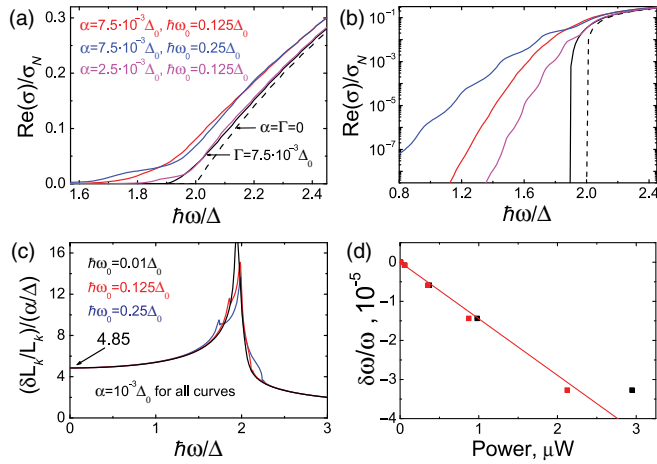


FIG. 2. (a) The real part of the conductivity  $\text{Re}\sigma(\omega)$  with the same values of depairing parameters  $\alpha$ ,  $\Gamma$  and signal frequencies  $\omega_0$  as for the densities of states in Figs. 1(b) and 1(c). (b) The real part of the conductivity on a logarithmic scale to highlight the effect of the low-lying states. The color coding of (a) applies. (c) The changes in the imaginary part of the conductivity due to the microwave power for values of  $\alpha$  and  $\hbar\omega_0$  as indicated in the figure. (d) The observed shift of the resonant frequency of a superconducting resonator as a function of the power of the microwave radiation. The red line shows the shift of the resonant frequency as a function of the internal power in the resonator, calculated using Eq. (7). Black squares represent the experimental data based on data analysis with a simple Lorentzian resonance curve [12]. The red squares correspond to the experimental data [12], based on an improved analysis by taking into account the nonlinearity of the kinetic inductance.

approach introduced by Eliashberg [11]. However, it was found that, at the lowest temperatures, it is not possible to account for the shift in resonant frequency. The observed shift was stronger than one would expect on the basis of the shift in quality factor, which is dominated by the redistribution of quasiparticles over the energies. In the present theoretical analysis, the new ingredient is the modification of the superconducting ground state in the presence of microwaves. Therefore, we focus on the change of resonant frequency with microwave power, which is a measure of the change in kinetic inductance, i.e., the Cooper-pair density, whereas the quality factor is a measure of the losses due to the quasiparticles. The kinetic inductance is given by the imaginary part of the conductance, which is shown in Fig. 2(c) for a selection of  $\alpha$ 's and  $\omega_0$ 's. Substitution of the calculated Green's functions into the imaginary part of Eq. (6) gives, for low values of  $\alpha$ , low frequencies [corresponding to the condition (1)], and low temperatures  $T \ll T_c$  a simple linear relation between  $\alpha$  and the shift of kinetic inductance  $\delta L_k$  with respect to its value without the rf field  $L_k$ :

$$\delta L_k/L_k \cong 4.85\alpha/\Delta_0. \quad (7)$$

To facilitate the comparison of Eq. (7) with the recent experiments [12], we rewrite Eq. (7) in the quantities which were measured in the experiment: the relative shift of the resonant frequency  $\delta\omega/\omega \cong -2.42P_{\text{in}}/P_0$ . Here,  $P_{\text{in}}$  is the (internal) power of the microwave radiation in the resonator and  $P_0 = 2\pi Z_0 N_0 \Delta_0^3 w^2 d^2 / (\hbar\rho)$  is the material parameter which relates the critical current to power, with  $Z_0$  being the impedance of the coplanar waveguide,  $\rho$  the normal state resistivity of the superconducting resonator (Al), and  $w$  and  $d$  the width and the thickness of the superconducting strip, respectively. This conversion was done under the assumption of a uniform distribution of the supercurrent across the strip.

In Fig. 2(d) we show the shift of the resonant frequency as a function of microwave power calculated using Eq. (7) as the red line. In the same figure, the black squares correspond to the experimental data of de Visser *et al.* [12], determined for a resonant frequency of 5.3 GHz, well above  $kT$  (64 mK) and well below  $\Delta$  (2.2 K). We include the experimental data as black squares, which agree very well with the theoretical prediction, except at the highest power. However, in the analysis of the measured resonance curves, the nonlinearity in the kinetic inductance was not taken into account. By including the nonlinearity of the kinetic inductance in the analysis of the measured resonance curves following Swenson *et al.* [33], the black data points from Ref. [12] were replaced by the red data points [Fig. 2(d)], which agrees very well with the theoretical prediction of Eq. (7) without using fitting parameters.

These results also have implications for the density of thermal quasiparticles for a given temperature. Obviously, the reconfiguration of the ground state in the presence of microwave power reduces the condensation energy, and therefore we expect for a given bath temperature a rapid increase in the number of quasiparticles. The number of excess quasiparticles  $N_{qp}$  can be measured by further detailed studies of their fluctuations in a superconducting strip [13]. We note that these other observables of the experiment [12] (the number of quasiparticles, the recombination time, the quality factor), which more dominantly depend on  $f(E)$  as compared to  $\delta\omega/\omega$ , are still dominated by a redistribution of the quasiparticles due to microwave absorption (Eliashberg [11]). In contrast,  $\delta\omega/\omega$  is a measure of the Cooper-pair current.

A direct test of the present theory would be a measurement of the density of states by tunneling. In such an experiment, the time-dependent electric field should only be in one of the electrodes parallel to the tunnel junction and not across the tunnel junction. This requirement is to avoid that the dc tunneling process itself is controlled by an additional ac electric field across the tunnel barrier, the well-known effect of photon-assisted tunneling [28,29]. A dc electric field should be across the tunnel junction to probe the electrodes with the tunneling current. The device could, in practice, be made as a transmission line. A

solution for such an experiment has been implemented by Horstman and Wolter [34,35], which is very feasible with the current level of technology. Instead of the nonequilibrium distribution function, one should extract the density of states, similarly to the experiment for dc pairing carried out by Anthore *et al.* [2].

In summary, we have presented a theoretical analysis of the quantum depairing effect by a microwave field for a long thin strip of a diffusive *s*-wave superconductor. We have demonstrated that the density of states loses a sharp peak at the gap energy and acquires features at the photon points  $\Delta \pm n\hbar\omega_0$ . Also, we have shown that the density of states develops an exponential-like tail in the subgap region. Both phenomena are in strong contrast to the case of depairing by a dc current. We have demonstrated that the predicted effect is responsible for the shift of the resonant frequency of an Al superconducting resonator by microwave power [12].

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\*av.semyonov@mpgu.edu

†igor-devyatov@yandex.ru

‡T.M.Klapwijk@tudelft.nl

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