

# Incoherence and enhanced magnetic quantum oscillations in the mixed state of a layered organic superconductor \*

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## Abstract

We present a theory which is able to explain enhanced magnetic quantum-oscillation amplitudes in the superconducting state of a layered metal with incoherent electronic transport across the layers. The incoherence acts through the deformation of the layer-stacking factor which becomes complex and decreases the total scattering rate in the mixed state. This novel mechanism can compensate the usual decrease of the Dingle factor below the upper critical magnetic field caused by the intralayer scattering.

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In recent years it has been questioned whether the electronic properties of layered quasi-two-dimensional (2D) metals can be described within the usual fundamental concept of an anisotropic three-dimensional Fermi liquid [1].

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Well-studied examples, besides the cuprate superconductors and quasi-one-dimensional organic metals, are organic conductors of the type (BEDT-TTF)<sub>2</sub>X, where BEDT-TTF stands for bisethylenedithio-tetrathiafulvalene and X for a monovalent anion. This class of materials displays a number of unique properties such as an unconventional electronic interlayer transport, clear deviations of their magnetic quantum oscillations [2] from the standard three-dimensional (3D) Lifshitz–Kosevich theory [3], and puzzling features in the superconducting mixed state. After the first observation of the de Haas–van Alphen (dHvA) oscillations in the mixed state of a layered superconductor [4] it was firmly established that these oscillations are damped below the upper critical field  $B_{c2}$ . This damping was explained by several mechanisms reviewed in [5]. A recent observation that both the dHvA and the Shubnikov–de Haas (SdH) amplitudes are enhanced in the mixed state of the layered organic superconductor  $\beta''$ -(BEDT-TTF)<sub>2</sub>SF<sub>5</sub>CH<sub>2</sub>CF<sub>2</sub>SO<sub>3</sub> ( $\beta''$  salt in the following) [6] was a real surprise since it is in a sharp contrast to all experiments and theories known so far [5].

Theoretically, it is assumed that the quasiparticle scattering by the “vortex matter” just below the upper critical field,  $B_{c2}$ , is the main mechanism of the damping in this region yielding the Dingle-like additional damping factor  $R_s = \exp(-2\pi/\Omega\tau_s)$ . Here,  $\Omega$  is the cyclotron frequency and  $\hbar/\tau_s$  is the sum of the two terms:  $\hbar/\tau_{s1} = \Delta^2(\pi/\mu\hbar\Omega)^{1/2}$ , due to the intralayer scattering at vortices [7–9], and  $\hbar/\tau_{s2} = \beta\Delta^2/\mu\Omega\tau_0$ , which takes into account the scattering by impurities and vortices within the layers [10]. ( $\tau_s$  is the scattering time,  $\mu$  is the chemical potential,  $\Delta$  is the superconducting order parameter which just below  $B_{c2}$  is less than the Landau-level separation  $\Delta < \hbar\Omega$ , and  $\beta \simeq 1$ .)

The additional factor  $R_s$  describes the extra damping of the dHvA and SdH amplitudes only due to the intralayer scattering in 2D conductors. Within this concept, however, the oscillation-amplitude enhancement in the  $\beta''$  salt is not explicable. Anomalous dHvA oscillations have also been observed in YNi<sub>2</sub>B<sub>2</sub>C [11, 12]. These oscillations persist down to the surprisingly low field  $0.2B_{c2}$  [12]. A Landau-quantization scheme for fields well below  $B_{c2}$  in the periodic vortex-lattice state was developed in [13] and for a model with an exponential decrease of the pairing matrix elements in [10]. The observed recovery of the dHvA amplitudes for  $B \ll B_{c2}$  in YNi<sub>2</sub>B<sub>2</sub>C was explained by the enhancement of the special vortex-lattice factor depending on the Landau bands which become narrower when the vortex-lattice grows thinner [10].

The anomalous magnetic quantum oscillations in the mixed state of the  $\beta''$  salt are much more mysterious and poses the question on the peculiarity

of this material. The  $\beta''$  salt is the only one among the known superconductors which (i) displays enhanced magnetic quantum-oscillation amplitudes in the mixed state and (ii) has no 3D Fermi surface (FS), i.e., exhibits an incoherent electronic transport across the layers [14]. The incoherence means that the electronic properties of this layered quasi-2D metal cannot be described within the usual fundamental concept of an anisotropic 3D FS. Nonetheless, magnetic oscillations due to the 2D FS survive [1].

Here, we consider a new mechanism for the quasiparticle scattering that goes beyond the usual 2D consideration of *intralayer* scattering by taking into account the *interlayer*-hopping contribution to the total scattering rate. This can explain an oscillation-amplitude enhancement in the superconducting state for layered conductors with incoherent hopping across the layers. The clear physical picture behind this mechanism is as follows. The incoherence, or disorder in the direction perpendicular to the layers, hampers the electron hopping between neighboring layers. This enhances the scattering at impurities within the layers since electrons on a Landau orbit interact with the same impurities many times before a hopping to the neighboring layer occurs. In the superconducting state long-range order establishes across the layers which allows quasiparticles to escape the *intralayer* multiple scattering by Josephson tunneling between the layers. This mechanism reduces the scattering rate by impurities and enhances the Dingle factor in the superconducting state. For the  $\beta''$  salt this effect most likely plays the dominant role. For the coherent case, there is no interlayer scattering and the electrons (quasiparticles) can move freely across the layers both in the normal and the superconducting state which render the above mechanism much less effective. Numerically, the effect is described by the layer-stacking factor [Eq. (3)] which itself contains a Dingle-like exponent in the case of incoherent interlayer hopping [15]. Superconductivity restores the coherence across the layers by renormalizing the hopping integrals [16]. This reduces the interlayer scattering and enhances the oscillation amplitudes.

In case the momentum across the layers is not preserved, the electron interlayer hopping can be described in terms of an energy  $\varepsilon$  that is distributed with the density of states (DOS)  $g(\varepsilon)$ . The energy spectrum of a layered conductor in a perpendicular magnetic field is, therefore, given by  $E_n(\varepsilon) = \hbar\Omega(n+1/2) + \varepsilon$  [15]. The total DOS then follows from the standard Green-function definition

$$N(E) = \frac{1}{2\pi^2 l^2} \text{Im} \sum_{n=0}^{\infty} \int d\varepsilon \frac{g(\varepsilon)}{E - E_n - \varepsilon - \Sigma_n(E)}, \quad (1)$$

where  $\Sigma_n(E)$  is the average self energy corresponding to the  $n$ th Landau

level and  $l = (\hbar c / eB)^{1/2}$  is the magnetic length. For large energies and large  $n \approx E / \hbar \Omega \gg 1$  (relevant for the magnetic quantum oscillations here) the self energy is independent of the index  $n$ , and the summation in Eq. (1) by use of the Poisson's formula yields

$$\frac{N(E)}{N(0)} = 1 + 2\text{Re} \sum_{p=1}^{\infty} (-1)^p R_D(p, E) I_p \exp\left(\frac{2\pi i p E}{\hbar \Omega}\right), \quad (2)$$

where  $N(0)$  is the 2D electron-gas DOS and the function

$$R_D(p, E) = \exp(-2\pi p |\text{Im}\Sigma(E)| / \hbar \Omega)$$

generalizes the Dingle factor to the case of an energy-dependent self energy  $\Sigma(E)$ . The layer-stacking factor in Eq. (2),

$$I_p = \int d\varepsilon g(\varepsilon) \exp\left(\frac{2\pi i p \varepsilon}{\hbar \Omega}\right), \quad (3)$$

is an important factor in the theory of magnetic quantum oscillations in normal and superconducting layered systems [10, 17]. It describes contributions to the oscillations coming from the interlayer hopping. If the stacking is irregular,  $I_p$  becomes complex and contains a Dingle-like exponent [15].

The inverse scattering time  $1/\tau(E) = |\text{Im}\Sigma(E)|/\hbar$  in the self-consistent Born approximation (SCBA) was found to be proportional to the total DOS [18–20]. Accordingly, the relation  $N(E)/N(0) = \tau_0/\tau(E)$  holds, where  $\tau_0$  is the intralayer scattering time [21, 22]. Substituting this into Eq. (2) leads to an equation for  $\tau(E)$  showing that it oscillates as a function of  $1/B$ .

In case the highly anisotropic electronic system has a 3D FS, the DOS related to the interlayer hopping is symmetric,  $g(\varepsilon) = g(-\varepsilon)$ , and for nearest-neighbor hopping becomes  $g(\varepsilon) = \pi^{-1}(4t^2 - \varepsilon^2)^{-1/2}$ . The corresponding layer-stacking factor then is given by  $I_p = J_0(4\pi t p / \hbar \Omega)$ . This Bessel function oscillates as a function of  $1/B$  which is just another way to describe the well-known bottleneck and belly oscillations of a corrugated 3D FS. Oscillating corrections to the Ginzburg–Landau expansion coefficients caused by the factor  $J_0(4\pi t p / \hbar \Omega)$  were also calculated in [23].

For the incoherent case, on the other hand, the translation invariance across the layers is lost. The irregular *inter*layer hopping means that the DOS deviates from the function  $g(\varepsilon) = \pi^{-1}(4t^2 - \varepsilon^2)^{-1/2}$  and loses the symmetry  $g(\varepsilon) = g(-\varepsilon)$  which implies that  $\text{Im}I_p \neq 0$ . As will be shown below, this results in a special contribution to the electron scattering time. Below  $B_{c2}$ , this may lead to a suppression of the scattering rate acting against the

known intralayer damping mechanisms of the quantum oscillations in the mixed state [5]. However, this contribution vanishes if the hopping between the layers preserves the interlayer momentum leading to a corrugated 3D FS cylinder. This is an important point in our consideration.

Using Eq. (2) and  $N(E)/N(0) = \tau_0/\tau(E)$  we write  $\tau(E)^{-1}$  as a sum of the coherent (symmetric) and incoherent (asymmetric) terms:

$$\tau(E)^{-1} = \tau(E)_s^{-1} - \tau(E)_a^{-1}, \quad (4)$$

$$\frac{\tau_0}{\tau_s} = 1 + 2 \sum_{p=1}^{\infty} (-1)^p R_D(p, E) \operatorname{Re} I_p \cos \left( \frac{2\pi p E}{\hbar \Omega} \right), \quad (5)$$

$$\frac{\tau_0}{\tau_a} = 2 \sum_{p=1}^{\infty} (-1)^p R_D(p, E) \operatorname{Im} I_p \sin \left( \frac{2\pi p E}{\hbar \Omega} \right). \quad (6)$$

With the help of the summation rule

$$S(\lambda, \delta) = \sum_{p=-\infty}^{\infty} (-1)^p e^{-|p|\lambda} \cos p\delta = \frac{\sinh \lambda}{\cosh \lambda + \cos \delta} \quad (7)$$

one can rewrite Eqs. (5) and (6) in the integral form

$$\frac{1}{\tau_{s(a)}} = \frac{1}{\tau_0} \int d\varepsilon g_{s(a)}(\varepsilon) S[\lambda, \delta(E, \varepsilon)]. \quad (8)$$

Here  $g_s(\varepsilon) = g_s(-\varepsilon)$  is the symmetric and  $g_a(\varepsilon) = -g_a(-\varepsilon)$  is the antisymmetric part of the DOS  $g(\varepsilon)$ ,  $\lambda(E) = 2\pi/\Omega\tau$ , and  $\delta(E, \varepsilon) = 2\pi(E - \varepsilon)/\hbar\Omega$ . The SCBA, as well as Eqs. (4)-(8), are valid not only for point-like impurities but also for a smooth random potential provided its correlation radius is less than the Larmor radius, which holds for large  $n$  [20]. One can see from Eqs. (4)-(8) that, *in general*, the incoherent contribution,  $-\tau(E)_a^{-1}$ , to the total scattering rate,  $\tau(E)^{-1}$ , is essential. The integral equation for  $\tau(E)^{-1}$  is very complicated and can be solved only perturbatively in the case  $\lambda \gg 1$ . In the limit  $\lambda \rightarrow \infty$ , when  $S(\lambda, \delta) \rightarrow 1$ , we have  $\tau_s^{-1} = \tau_0^{-1}$  and  $\tau_a^{-1} = 0$ . For finite but large  $\lambda$  the parameter  $R_D(p, E) = e^{-p\lambda} \ll 1$ . Even if  $\Omega\tau \simeq 1$ , the quantity  $e^{-\lambda} \ll 1$  and Eqs. (5) and (6) are just a series expansion in powers of the small parameter  $e^{-\lambda}$ . Eq. (7) shows the convergence of this series for any  $\lambda > 0$  allowing a perturbative solution. The perturbative terms oscillate as a function of  $E$  and can be written as the series  $\tau^{-1}(E) = \tau_0^{-1}[1 + X_1 + X_2 + O(e^{-3\lambda_0})]$ , with  $X_1 \propto e^{-\lambda_0}$ ,  $X_2 \propto e^{-2\lambda_0}$ ,

and  $\lambda_0 = 2\pi/\Omega\tau_0$ . The first nonzero correction averaged over an oscillation period is proportional to  $\overline{X_1 + X_2} = -2\lambda_0 e^{-2\lambda_0} |I_1|^2$  and yields

$$\frac{1}{\bar{\tau}} = \frac{1}{\tau_0} \left[ 1 - \frac{4\pi}{\Omega\tau_0} (R_D^0)^2 (\text{Re}I_1^2 + \text{Im}I_1^2) \right]. \quad (9)$$

The Dingle factor  $R_D^0 = \exp(-2\pi/\Omega\tau_0)$  is a small parameter in our perturbative solution. The term  $\text{Im}I_1^2$  in Eq. (9) appears due to the incoherence.

It was established that in the  $\beta''$  salt electron hopping across the layers is most probably incoherent, i.e., the momentum perpendicular to the layers is not preserved and there is no 3D FS [14]. The reason for this remarkable feature is unknown so far. It might be that some kind of disorder, such as different spatial configurations in the extraordinary large and complex anion-molecule layer, may induce random hopping integrals, by analogy with intercalated layered compounds [15]. Furthermore, the  $\beta''$  salt is the only material so far studied (not only among the BEDT-TTF salts)

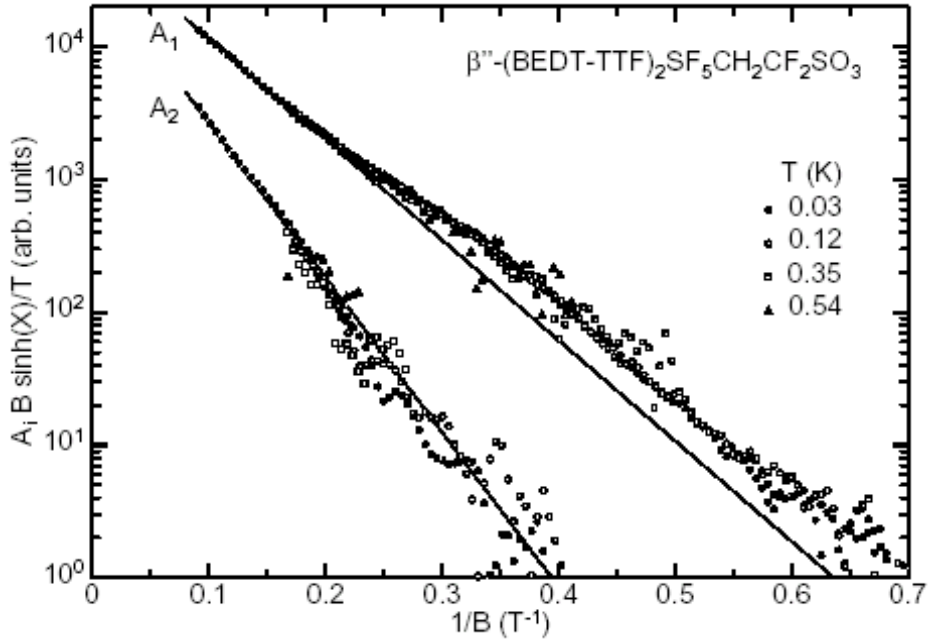


Figure 1: Dingle plot of the fundamental ( $A_1$ ) and the second harmonic ( $A_2$ ) dHvA amplitudes of the  $\beta''$  salt extracted from modulation-field data for different temperatures. The solid lines are fits to the data above 6 T.

which displays an enhancement of the magnetic quantum-oscillation amplitude in the superconducting state [6].

This important result is summarized in Fig. 1. In this Dingle plot for a 2D metal the amplitudes  $A_1$  of the fundamental dHvA frequency ( $F \approx 198$  T) and  $A_2$  of the second harmonic ( $2F$ ) are normalized by  $B \sinh(X)T^{-1}$  and plotted on a logarithmic scale as a function of  $1/B$ , with  $X = 2\pi^2 k_B m_c T / e\hbar B$  and  $m_c$  the effective cyclotron mass (see [6] for more details). Upon entering the superconducting state the oscillation amplitude  $A_1$  is enhanced compared to the normal-state dependence (solid lines). For the second harmonic  $A_2$  neither an additional damping nor an enhancement is observed [24].

In the superconducting state long-range order across the layers evolves through the renormalization of the hopping integrals [16]. This can be understood as follows. The quasiparticle hopping between the layers is an independent degree of freedom with respect to the in-plane Landau quantization. In the normal state the Green-function equation related to the interlayer hopping is given by

$$\sum_m [(\varepsilon - \varepsilon_i)\delta_{im} - t_{im}]G_{ij}^0(\varepsilon) = \delta_{ij}, \quad (10)$$

where the electron energy in the layer  $\varepsilon_i$  and the hopping integrals  $t_{im} = t_i(\delta_{i,m+1} + \delta_{i,m-1})$  are assumed to depend on the layer indices for the sake of generality. In the superconducting state the order parameters in the layers,  $\Delta_i$ , become nonzero and the Gor'kov equation for the Green functions  $G_{ij}$  can be written as [16]

$$\sum_m [(\varepsilon - \varepsilon_i)\delta_{im} - \hat{t}_{im}]G_{ij}(\varepsilon) = \delta_{ij}, \quad (11)$$

$$\hat{t}_{im} = t_{im} - \Delta_i G_{im}^0(-\varepsilon)\Delta_m^*. \quad (12)$$

The star means the complex conjugate. When comparing Eq. (10) with Eq. (11) it is seen that in the superconducting state the effective hopping integrals, given by Eq. (12), become nonzero not only for next-nearest-neighbor hopping. In that case the nonvanishing retarded Green-function components  $G_{im}^0(-\varepsilon)$  result in nonzero  $\hat{t}_{im}$  for electron hopping between arbitrary sites  $i$  and  $m$ . In fact, Eq. (12) simply reflects how the superconducting long-range order establishes across the layers. The complex order parameters in the layers  $\Delta_i = |\Delta_i| \exp(i\varphi)$  appear and result in interlayer (intrinsic) Josephson coupling [16]. In the absence of Josephson currents the order parameter

can be chosen to be real and independent of the layer index  $\Delta_i = |\Delta_i|$ . The correction to the DOS due to this mechanism is

$$\delta g(\varepsilon) = -\Delta^2 \sum_{ij} \frac{\partial g(\varepsilon)}{\partial t_{ij}} G_{ij}^0(-\varepsilon). \quad (13)$$

The second effect we have to take into account is caused by the vortex matter in the mixed state. Here, the vortices are disordered for fields slightly below  $B_{c2}$ . They convert the degenerated Landau levels into asymmetric Landau bands as was shown in Refs. [25, 26]. Thus, in the mixed state the quasiparticle tunneling between the layers implies the quantum transition between these Landau-band states which results in the additional contribution to the simple nearest-neighbor DOS

$$\delta g_{\Delta}(\varepsilon) = \Delta^2 \left( \frac{\partial g(\varepsilon)}{\partial \Delta^2} \right)_{\Delta=0}. \quad (14)$$

The total correction to the DOS in the mixed state can be written as  $\delta g_{\text{tot}}(\varepsilon) = \Delta^2 G(\varepsilon)$ , where  $G(\varepsilon)$  is directly defined by Eqs. (13) and (14). Inserting  $\delta g_{\text{tot}}(\varepsilon)$  into Eq. (8) and averaging over a period in  $E$  results in

$$\delta \left( \frac{1}{\tau} \right) = \frac{\Delta^2}{\tau_0} \int d\varepsilon G(\varepsilon) \overline{S(\lambda, \delta(E, -\varepsilon))} = \frac{\Delta^2}{\tau_0} \gamma. \quad (15)$$

Since the DOS is normalized ( $\int d\varepsilon g(\varepsilon) = 1$ ), the function  $G(\varepsilon)$  satisfies the condition  $\int d\varepsilon G(\varepsilon) = 0$ . This means that it is alternating in sign and  $\gamma$  might be negative because  $\overline{S(\lambda, \delta(E, -\varepsilon))} > 0$ . The studied system is too complex to calculate  $\gamma$  in general. In the limit  $\lambda \rightarrow \infty$  this coefficient vanishes since  $\overline{S(\lambda, \delta(E, -\varepsilon))} \rightarrow 1$ . It is instructive to consider a correction to the scattering rate in Eq. (9) in the mixed state. The variation of the layer-stacking factor  $\delta I_1$  is given by Eq. (3) with the DOS replaced by  $\delta g_{\text{tot}}(\varepsilon)$ . The broadening of the Landau levels, caused by  $\delta g_{\text{tot}}(\varepsilon)$ , is of the order of the width of this function and much less than  $\hbar\Omega$  in order to observe the oscillations. Therefore, in the first approximation  $\text{Re}\delta I_1 \approx 0$  and  $\text{Im}\delta I_1 = \Delta^2 \int d\varepsilon G(\varepsilon) \sin(\frac{2\pi\varepsilon}{\hbar\Omega}) \approx \Delta^2 (\frac{2\pi\langle\varepsilon\rangle}{\hbar\Omega})$ , where  $\langle\varepsilon\rangle = \int d\varepsilon G(\varepsilon)\varepsilon$ . (For  $G(\varepsilon) = -G(-\varepsilon)$ ,  $\text{Re}\delta I_1$  is zero exactly.) Thus, the correction to the scattering rate in the mixed state near  $B_{c2}$ , caused by the interlayer-hopping mechanism, is given by

$$\delta \left( \frac{1}{\tau} \right) = -\Delta^2 \frac{2\text{Im}I_1}{\tau_0} \left( \frac{4\pi}{\Omega\tau_0} \right) (R_D^0)^2 \left( \frac{2\pi\langle\varepsilon\rangle}{\hbar\Omega} \right). \quad (16)$$



For  $\langle \varepsilon \rangle \text{Im}I_1 > 0$ , this gives a decrease in the scattering rate. Note that the latter is nonzero only if the system is incoherent in the normal state and  $\text{Im}I_1 \neq 0$ . This strongly supports the relevance of this mechanism for the  $\beta''$  salt, since only this organic metal displays both incoherence in the normal state and an enhancement of the SdH and dHvA amplitudes in the mixed state.

Thus, the overall effect superconductivity has on  $R_s$  is determined by the balance between positive and negative contributions to the scattering rate. The already mentioned positive contribution from the intralayer scattering at vortices and defects is

$$\frac{\hbar}{\tau_s} = \Delta^2 \left[ \left( \frac{\pi}{\mu \hbar \Omega} \right)^{1/2} + \frac{\beta}{\mu \Omega \tau_0} \right]. \quad (17)$$

The new additional interlayer mechanism we discuss here results in a negative contribution given by  $\delta \left( \frac{1}{\tau} \right) = \frac{\Delta^2}{\tau_0} \gamma$  [Eq.(15)]. Since little is known about the DOS of the studied system, even in the normal state, we cannot calculate the coefficient  $\gamma$  quantitatively. However, contrary to Eq. (17), for this term the small factor  $1/\mu$  is absent, so that the overall correction to the scattering rate might be negative. The experimental facts [6] give us confidence that this is the case at least for the  $\beta''$  salt.

We conclude with a qualitative picture of the effect discussed here. The incoherence means that the hopping time between the layers  $\tau_z \approx \hbar/|t| \gg \tau_0$  so that an electron scatters many times within a layer before leaving it [1]. Here the quantity  $|t|$  is some averaged hopping integral that in the  $\beta''$  salt may be assumed to be the smallest parameter in energy. Indeed, experimentally  $|t|$  cannot be resolved in the  $\beta''$  salt reflecting the fact that the hopping integral is one of the smallest for all known 2D organic metals so far [14]. Consequently, even small spatial fluctuations of the hopping probability within and across the layers render the electron motion across the layers incoherent. On the other hand, for the evolution of superconductivity some interlayer (Josephson) coupling is vitally important. Long-range order establishes below  $B_{c2}$ , thereby renormalizing the hopping integral. According to Eq. (12), the renormalized  $\tau_z$  in the superconducting state may be estimated as  $\tau_z \approx \hbar/|t + \Delta^2/t|$ . For  $\Delta \gg |t|$  the hopping time is reduced considerably and becomes  $\tau_z \approx \hbar|t|/\Delta^2$ . The latter means that the quasiparticles spend less time within the (impurity-containing) layers decreasing the scattering rate and, consequently, enhancing the Dingle factor. In the  $\beta''$  salt this effect is strong because of the smallness of  $|t|$ . Thus, our mechanism relates the two unusual effects observed in the  $\beta''$  salt: the incoherent in-

terlayer hopping transport and the enhancement in the quantum-oscillation amplitudes in the mixed state.

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