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Quasiparticle Interference as a Direct Experimental Probe of Bulk Odd-Frequency Superconducting Pairing

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We show that quasiparticle interference (QPI) due to omnipresent weak impurities and probed by Fourier transform scanning tunneling microscopy and spectroscopy acts as a direct experimental probe of bulk odd-frequency superconducting pairing. Taking the example of a conventional s-wave superconductor under applied magnetic field, we show that the nature of the QPI peaks can only be characterized by including the odd-frequency pairing correlations generated in this system. In particular, we identify that the defining feature of odd-frequency pairing gives rise to a bias asymmetry in the QPI, present generically in materials with odd-frequency pairing irrespective of its origin.

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The key to finding new superconductors lies in understanding the nature of the Cooper pairs as they are the fundamental building blocks of the superconducting state. In particular, the symmetry of the Cooper pair wave function is crucial in determining the superconductor's stability, both to intrinsic constituents such as disorder and to external perturbations like magnetic field. The earliest known metallic superconductors have been successfully described by the Bardeen, Cooper, and Schrieffer (BCS) theory [1], which is built on the Cooper pair wave function having spin-singlet s-wave symmetry and also an even-frequency dependence or, equivalently, being even under the exchange of the relative time coordinate of the paired electrons. Even after the discovery of high-temperature superconductors, the overall paradigm of BCS theory of superconductivity has remained fairly successful, albeit using other spin and spatial symmetries [2,3]. However, there exist several properties in superconductor-ferromagnetic heterostructures [4–12], which can only be explained by the presence of Cooper pairs with a wave function that is odd in frequency [13–21].

Experimental evidence of odd-frequency pairing in superconductor-ferromagnetic heterostructures [4–9,22] has motivated other theoretical works, proposing the existence of odd-frequency pairs also in bulk systems without the need of heterostructures [23–35]. Additionally, bulk odd-frequency pairing has also been discussed theoretically [36–39] and experimentally [40] in the context of heavy fermions for pairing near quantum critical points. However, a major challenge has been the experimental verification of such theoretical proposals of bulk odd-frequency superconducting pairs. While experiments have been successful in identifying odd-frequency pairing in heterostructures [4,6–9,22], there exists still no proposal of an easily accessible experimental probe which clearly identifies odd-frequency Cooper pairs in bulk systems. The challenge mainly arises due to the fact that bulk odd-frequency pairs are generally accompanied by even-frequency pairs and it is difficult to experimentally disentangle their different characteristics. A few proposals still exist, such as using the Kerr effect [30] or a paramagnetic Meissner effect [5,9,18,19,22]. However, the Kerr effect additionally requires time-reversal symmetry breaking [41,42] and the Meissner effect has been shown to be unreliable in multiband superconductors since odd-frequency pairing can also generate a diamagnetic Meissner signal [33,43]. Additionally, neither of these tools directly detect the oddness in frequency, but instead rely on indirect effects. Another theoretical proposal has tried to provide a direct experimental detection scheme of bulk odd-frequency Cooper pairs using time- and angle-resolved photoelectron fluctuation spectroscopy [44], but involves technology clearly beyond the scope of even the most advanced existing facilities.

In this Letter we show that the already existing experimental technique of scanning tunneling microscopy (STM) or scanning tunneling spectroscopy (STS) can directly detect odd-frequency superconducting pairing. In particular, we use that weak nonmagnetic impurities in superconductors are ever-present and create charge density inhomogeneities, which result in quasiparticle scattering. The resulting interference patterns can be probed experimentally by Fourier transformed STM/STS in a technique commonly referred to as quasiparticle interference (QPI) [45–47]. QPI has long been an important experimental probe to determine various signatures of superconducting pairing [48–51], especially the pairing symmetry in high-temperature superconductors such as iron-based superconductors [52–55]. Here, by considering a prototype system of a conventional spin-singlet s-wave superconductor under applied magnetic field, we show that odd-frequency pairing, known to be present in such systems [56–58], produces two direct signatures in the peak structure of the QPI. First, we show that the peak positions of the local density of states (LDOS) change can only be identified accurately if odd-frequency pair correlations are incorporated. Second, and most remarkably, the LDOS change at positive and negative applied bias voltage results in different peak positions, and their separation is directly related to the presence of odd-frequency pairs. This bias asymmetry arises due to the defining feature of odd-frequency pairing, i.e., the superconducting pair correlations being odd in frequency or, equivalently, here in applied bias voltage.

Model.—To model a simple bulk superconductor with odd-frequency superconducting pair correlations we use a conventional spin-singlet s-wave superconductor under applied magnetic field described by the mean-field Hamiltonian:

$$H = \sum_{k,\sigma}(\xi_k + \sigma B)c^\dagger_{k\sigma}c_{k\sigma} + \sum_k \Delta_0 c_{-k\downarrow}c_{k\uparrow} + \text{H.c.} \quad (1)$$

where $c^\dagger_{k\sigma}$ ($c_{k\sigma}$) is the creation (annihilation) operator of an electron with spin σ and momentum k, ξ_k is the electron band dispersion, B is the magnetic field with the magnetic moment of the electron μ_0 taken to be unity, Δ_0 is the spin-singlet isotropic s-wave superconducting order parameter, and we have ignored an overall constant. For simplicity, we use the band dispersion of a square lattice: $\xi_k = -2t[\cos(k_x) + \cos(k_y)] - \mu$, where $t = 1$ is the energy unit. We only consider the Zeeman effect of the applied magnetic field in this Letter. An applied magnetic field can also affect the orbital motion of electrons and create vortices in a superconductor. However, in two-dimensional superconductors with the magnetic field applied in plane, the only relevant effect is the Zeeman effect. Furthermore, Δ_0 is obtained by the self-consistency relation, $\Delta_0 = -\sum_{k'} U\langle c^\dagger_{k'\uparrow}c^\dagger_{-k'\downarrow}\rangle$ with U being the effective attraction driving the superconducting order.

The Hamiltonian in Eq. (1) can be written in a matrix form using the Nambu basis $\Psi^\dagger = (c^\dagger_{k\uparrow}, c_{-k\downarrow})$ as, $H = \sum_k \Psi^\dagger \hat{H}\Psi$ with

$$\hat{H} = \begin{pmatrix} \xi_{k\uparrow} & \Delta_0 \\ \Delta_0 & -\xi_{-k\downarrow} \end{pmatrix}, \quad (2)$$

where now $\xi_{k\sigma} = \xi_k + \sigma B$ and Δ_0 is, without loss of generality, taken to be real. We diagonalize the Hamiltonian $\hat{H}$ and solve the self-consistent equation of Δ_0 iteratively. We also tune μ such that the average density of electrons $\rho = \sum_{k,\sigma}\langle c^\dagger_{k\sigma}c_{k\sigma}\rangle$ is kept fixed to the generic value 0.7. Our findings do not qualitatively depend on the choice of the average density. We use a large system size, $N = 1000 \times 1000$ and $U = 2.5$ for obtaining a significant gap Δ_0 to make the analysis clear. We have verified that all qualitative features remain for experimentally realistic gap sizes. For the given set of parameters, we find the BCS superconducting state to be stable for $B < 0.35$.

Odd-frequency pair correlations.—Different correlations of the Hamiltonian in Eq. (2) can be obtained by calculating the corresponding Green's function G, given by $G^{-1}(i\omega) = i\omega - \hat{H}$, where ω are fermionic Matsubara frequencies. Using the Hamiltonian in Eq. (2), we thus obtain the Green's function by inverting the 2×2 matrix $G^{-1}(i\omega)$. In particular, the superconducting pair correlations are given by the anomalous, off-diagonal, part of the Green's function,

$$G_{12}(i\omega) = F_k(i\omega) = F^e_k(i\omega) + F^o_k(i\omega), \quad (3)$$

where we analytically extract

$$F^e_k(i\omega) = \frac{-\Delta_0(\xi_{k\uparrow}\xi_{-k\downarrow} + \Delta_0^2 + \omega^2)}{D}, \quad (4)$$

$$F^o_k(i\omega) = \frac{i\omega\Delta_0(\xi_{k\uparrow} - \xi_{-k\downarrow})}{D}, \quad (5)$$

$$D = (\xi_{k\uparrow}\xi_{-k\downarrow} + \Delta_0^2 + \omega^2)^2 + \omega^2(\xi_{k\uparrow} - \xi_{-k\downarrow})^2. \quad (6)$$

Here we have divided the superconducting pair correlations into even-frequency, $F^e_k(i\omega)$, and odd-frequency, $F^o_k(i\omega)$, contributions, as clearly set by the frequency dependence in the numerators since the common denominator D is an even function of frequency. Moreover, we see directly that odd-frequency pairs exist as soon as B is finite, as expected with a triplet spin symmetry [56–58].

QPI theory.—QPI probes the change in the LDOS due to omnipresent weak nonmagnetic impurities. The LDOS in the presence of such impurities can be decomposed as $\rho(r,\omega) = \rho_0(\omega) + \delta\rho(r,\omega)$, where ρ_0 is the DOS of a homogeneous superconductor and $\delta\rho$ is the change due to impurities. The corresponding Fourier transformed quantity $\delta\rho(q,\omega)$ is written in terms of the Green's function as [49–53,55]

$$\delta\rho(q,\omega) = -\frac{1}{\pi} Im\left[\sum_k G(k,\omega)TG(k+q,\omega)\right]_{11}, \quad (7)$$

where $G(k,\omega)$ is obtained by analytically continuing $i\omega \to \omega + i\eta$ in the unperturbed $G(k,i\omega)$ and T is the T matrix [59] corresponding to the impurity. Assuming weak nonmagnetic impurities, $T = V_{\text{imp}}\tau_3$ [52], where τ_3 is the third

Pauli matrix in the Nambu basis and we set $V_{\rm imp} = 1$ to mimic weak impurities. This impurity treatment is sufficient as the superconducting properties or the ground state is not changed in an s-wave superconductor for weak nonmagnetic impurities due to Anderson's theorem [60]. Moreover, weak nonmagnetic impurities do not create additional local odd-frequency correlations [61], thus helping in unambiguous detection of bulk odd-frequency correlations. Strong or magnetic impurities may create additional bound states. However, experimentally such bound states are often easily isolated and subtracted from the signal obtained for QPI [54]. The quantity $\delta\rho(q,\omega)$ is also experimentally accessible by Fourier transforming the LDOS obtained using STM or STS at different bias voltages $V = \omega/e$, where e is the electron charge [52].

For weak nonmagnetic impurities, Eq. (7) becomes

$$\delta\rho(q,\omega) = -\frac{1}{\pi}\mathrm{Im}\Big[\sum_k G_{11}(k,\omega)G_{11}(k+q,\omega) - G_{12}(k,\omega)G_{21}(k+q,\omega)\Big],$$

$$= -\frac{1}{\pi}\mathrm{Im}\Big[\sum_k G_{11}(k,\omega)G_{11}(k+q,\omega) \tag{8a}$$

$$- F^e_k(\omega)F^e_{k+q}(\omega) - F^o_k(\omega)F^o_{k+q}(\omega) \tag{8b}$$

$$- F^e_k(\omega)F^o_{k+q}(\omega) - F^o_k(\omega)F^e_{k+q}(\omega)\Big], \tag{8c}$$

where $F^e_k(\omega)$ and $F^o_k(\omega)$ are analytically continued versions of $F^e_k(i\omega)$ and $F^o_k(i\omega)$, respectively. Looking at the expressions in Eq. (8), we can already make two key observations. First, $\delta\rho(q,\omega)$ directly access the pair correlations through the terms proportional to F^e_k and F^o_k. This is in sharp contrast to the homogeneous DOS $\rho_0(\omega) = -1/\pi\sum_k \mathrm{Im}[G_{11}(k,\omega)]$, which only captures the superconducting energy gap Δ_0 through G_{11} and can thus not probe the pair correlations $F^{e/o}_k$. Second, the expression Eq. (8) has no integration over ω. As a result, the contributions coming from the product of the even- and odd-frequency correlations are generically nonzero. These contributions are odd in ω by definition, in contrast to the contributions coming from so-called even-even $F^e_k(\omega)F^e_{k+q}(\omega)$ or odd-odd $F^o_k(\omega)F^o_{k+q}(\omega)$ correlations. Hence, we already here find that the presence of odd-frequency correlations generically, irrespective of its origin, influences $\delta\rho(q,\omega)$.

QPI results.—In order to identify the role of odd-frequency correlations in the LDOS change $\delta\rho(q,\omega)$, we compare $\delta\rho(q,\omega)$ with $\delta\rho_e(q,\omega)$, where $\delta\rho_e(q,\omega)$ is calculated altogether ignoring odd-frequency correlations and keeping only even-frequency correlations, i.e., $F^o_k(\omega)$ is set to zero in the expression in Eq. (8) to obtain $\delta\rho_e(q,\omega)$.

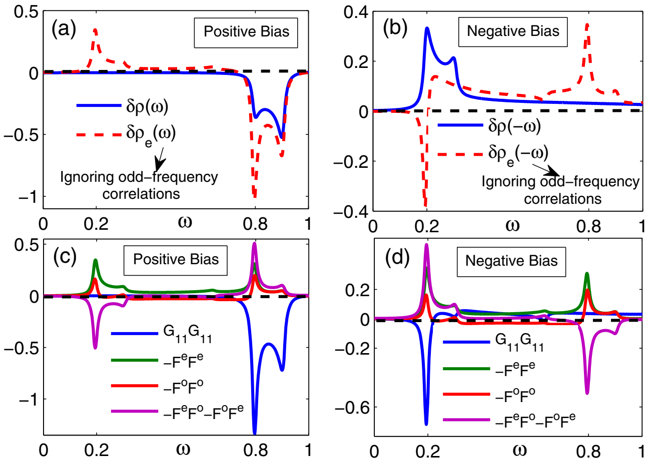

FIG. 1. LDOS change as a function of ω for $B = 0.3$ and $q = (1.37, 0)$, showing different peak structures. $\delta\rho$ and $\delta\rho_e$ for positive bias (a) and negative bias (b). Individual contributions of each term in Eq. (8) to $\delta\rho$ for positive bias (c) and negative bias (d). Black dashed line marks zero. Results are obtained for an artificial broadening $\eta = 0.01$.

We here present ω in units of t and the LDOS change in units of inverse t.

In Figs. 1(a) and 1(b) we show the LDOS changes $\delta\rho(q,\omega)$ and $\delta\rho_e(q,\omega)$ considering the system in Eq. (1) as a function of ω for a fixed $q = (1.37, 0)$ and $B = 0.3$. The choice of B is set to give significant odd-frequency correlations, as also illustrated later in Fig. 3. We show in the Supplemental Material (SM) [62] that the qualitative features obtained in Fig. 1 are indeed independent of the choice of q or B. Focusing on the positive bias ($\omega > 0$) in Fig. 1(a), we find that $\delta\rho(\omega)$ has a negative double-peak structure around $\omega = 0.8 = \Delta_0 + B$. When ignoring odd-frequency correlations, $\delta\rho_e(\omega)$ shows also an additional peak at $\omega = 0.2 = \Delta_0 - B$. The lack of this spurious peak in the full $\delta\rho(q,\omega)$ is the first evidence of the presence of odd-frequency correlations. If we look at the negative bias $\delta\rho_e(-\omega)$ in Fig. 1(b), we find a similar spurious peak at $\omega = 0.8$. The negative bias results show further qualitative differences between $\delta\rho$ and $\delta\rho_e$. Around $\omega = 0.2$, $\delta\rho(-\omega)$ has a positive double peak. In contrast, $\delta\rho_e(-\omega)$ has a sign change occurring near $\omega = 0.2$, such that $\delta\rho_e(-\omega) < 0$ for $\omega < 0.2$ and $\delta\rho_e(-\omega) > 0$ for $\omega > 0.2$. These results illustrate clearly that the LDOS change as a function of ω attains a different peak structure due to the presence of odd-frequency correlations.

The appearance of spurious peaks in $\delta\rho_e$ in contrast to the observable $\delta\rho$ for both positive and negative bias in Figs. 1(a) and 1(b) can be understood by looking at the individual components of $\delta\rho$ in Eq. (8). In Figs. 1(c) and 1(d) we show the contributions coming from the normal, diagonal, part of the Green's function $G_{11}G_{11} = -\mathrm{Im}[\sum_k G_{11}(k,\omega)G_{11}(k+q,\omega)]/\pi$, i.e., the term in Eq. (8a), and the anomalous or superconducting,

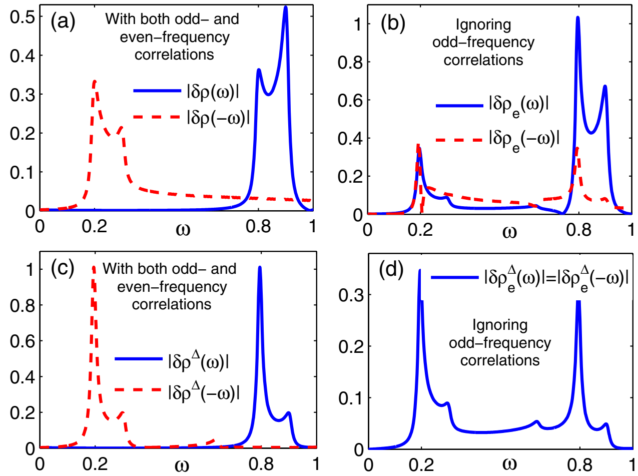

FIG. 2. Absolute value LDOS change as a function of ω for $B = 0.3$ and $q = (1.37, 0)$, showing bias asymmetry. Positive and negative bias values of $|\delta\rho|$ (a) and $|\delta\rho_e|$ (b), as well as the contributions only from superconductivity $|\delta\rho^\Delta(\pm\omega)|$ (c) and $|\delta\rho_e^\Delta(\pm\omega)|$ (d). Artificial broadening is same as in Fig. 1.

off-diagonal, part of the Green's function $F^{e/o}F^{e/o} = -\mathrm{Im}[\sum_k F_k^{e/o}(\omega)F_{k+q}^{e/o}(\omega)]/\pi$, i.e., the terms in Eqs. (8b) and (8c). In particular, we note that the contribution from $F^eF^o + F^oF^e$ exactly cancels $F^eF^e + F^oF^o$ at $\omega = 0.2$ for positive bias in Fig. 1(c) and at $\omega = 0.8$ for negative bias in Fig. 1(d). These exact cancellations can be easily seen by comparing the total $\delta\rho$ in Figs. 1(a) and 1(b) with the normal part $G_{11}G_{11}$ in Figs. 1(c) and 1(d). Moreover, $G_{11}G_{11}$ do not feature any peaks at these ω values. As a result, it is only in the presence of odd-frequency correlations, i.e., when $F^o \neq 0$, that there are no spurious peaks at $\omega = 0.2$ for positive bias and $\omega = 0.8$ for negative bias in $\delta\rho$.

Further looking for signatures of odd-frequency correlations, we reemphasize that the defining feature of odd-frequency correlations is that they are odd in frequency or, equivalently, energy bias. Therefore, we particularly want to explore the possibility of a bias asymmetry in the LDOS change. For this purpose, we plot the absolute values $|\delta\rho(\omega)|$ and $|\delta\rho(-\omega)|$ in Fig. 2(a), and for comparison $|\delta\rho_e(\omega)|$ and $|\delta\rho_e(-\omega)|$ in Fig. 2(b), again using a fixed $q = (1.37, 0)$ with $B = 0.3$. In Fig. 2(a), $|\delta\rho(\omega)|$ shows the double peak centered around $\omega = 0.8$, whereas the double peak of $|\delta\rho(-\omega)|$ is centered around $\omega = 0.2$. This illustrates directly a clear asymmetry in the peak positions of $|\delta\rho(\omega)|$ and $|\delta\rho(-\omega)|$ when odd-frequency correlations are appropriately included. In comparison, the LDOS change if ignoring odd-frequency correlations is shown in Fig. 1(b). It is evident from Fig. 1(b) that both $|\delta\rho_e(\omega)|$ and $|\delta\rho_e(-\omega)|$ have peaks around both $\omega = 0.2$ and $\omega = 0.8$. Thus, there is no asymmetry in the positions of the peaks of $|\delta\rho_e(\omega)|$ and $|\delta\rho_e(-\omega)|$. As a consequence, the bias asymmetry in the peak positions of $|\delta\rho(\omega)|$ and $|\delta\rho(-\omega)|$ becomes a clear and robust experimental signature of the presence of odd-frequency correlations. However, we still note that the heights of the peaks for positive and negative bias are asymmetric in both Figs. 1(a) and 1(b). To analyze this peak height asymmetry, in Figs. 1(c) and 1(d) we plot the LDOS change after subtracting the contribution coming from the normal part of the Green's function, resulting in the isolation of the contribution from superconducting correlations, $\delta\rho^\Delta = \delta\rho - G_{11}G_{11}$. If the odd-frequency correlations are ignored, as is shown in Fig. 1(d), then the positive and negative bias results are exactly identical, i.e., $|\delta\rho_e^\Delta(\omega)| = |\delta\rho_e^\Delta(-\omega)|$. This is an expected result since the only contribution to $\delta\rho^\Delta$ comes from F^eF^e, which is an even function in frequency. Moreover, once the odd-frequency correlations are appropriately included, we find that $\delta\rho^\Delta(\omega)$ and $\delta\rho^\Delta(-\omega)$ peak at $\omega = 0.8$ and $\omega = 0.2$, respectively, and the peak heights are in fact identical. Thus, the asymmetry in the peak heights in Figs. 2(a) and 2(b) is stemming from the contribution of the normal part of the Green's function. As a consequence, it is the bias asymmetry in the peak positions that is the decisive tool to prove odd-frequency pairing, while the heights are varying with the normal, nonsuperconducting properties. In the SM we show that these are robust features, not dependent on the choice of q or B. Another striking feature is that the bias asymmetry only appears in the LDOS change, or QPI, and not in any spatially averaged tunneling measurements, as the average DOS does not capture the superconducting correlations, as mentioned earlier. Hence, averaged tunneling measurements see two symmetric peaks at $\omega = \Delta_0 \pm B$ for both positive and negative bias [58,65].

In order to further justify the bias asymmetry originating from odd-frequency pair correlations, we show the dependence of the odd-frequency correlations on the applied magnetic field B in the inset of Fig. 3. Here we plot the momentum integrated values $F^o(\omega) = \sum_k |ImF_k^o|$ for three different values of B. F^o is clearly zero at $\omega = 0$ and peaks to a maximum value $F^o_{\max}$ at a finite ω. With increasing B, $F^o_{\max}$ also increases. This increase can easily be understood by looking at the expression for $F_k^o(i\omega)$ in Eq. (5). The numerator of Eq. (5) depends linearly on $\xi_{k\uparrow} - \xi_{k\downarrow} = 2B$ and it also primarily decides the maximum value $F^o_{\max}$. To relate these odd-frequency pair correlations with the bias asymmetry we uncovered in Fig. 2, we first note that the distance $\Delta\omega$ between the peaks of $\delta\rho^\Delta(\omega)$ and $\delta\rho^\Delta(-\omega)$ is always equal to $2B$, since $\delta\rho^\Delta(\omega)$ peaks at $\omega = \Delta_0 + B$ and $\delta\rho^\Delta(-\omega)$ peaks at $\omega = \Delta_0 - B$. To numerically substantiate this result, we show in the main panel of Fig. 3 the relation between $\Delta\omega$ and $F^o_{\max}$ obtained at multiple different B. Importantly, as seen in Fig. 3, $\Delta\omega$ is directly correlated to $F^o_{\max}$. This correlation establishes the direct connection of the odd-frequency correlations and the bias asymmetry in the LDOS change. The correlation plot in Fig. 3 is not quite linear because the B dependence of $F^o_{\max}$

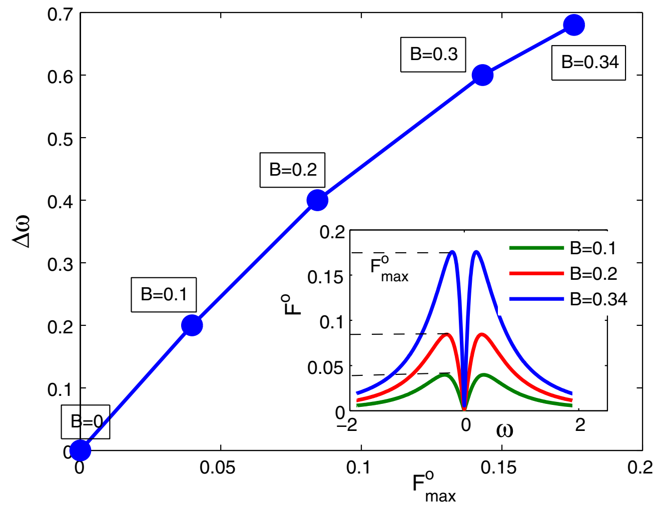

FIG. 3. Correlation between the energy separation $\Delta\omega$ between the peaks of $|\delta\rho^{\Delta}(\omega)|$ and $|\delta\rho^{\Delta}(-\omega)|$ and maximum value of the momentum averaged odd-frequency pair correlations $F^{o}_{\max}$ for multiple different B (indicated at each point). Inset: F^{o} for different values of magnetic field B, with $F^{o}_{\max}$ values indicated by dashed black lines.

is not exactly linear due to also a B dependence in the denominator in Eq. (5).

Concluding remarks.—We showed that odd-frequency superconducting pair correlations can be directly probed by the quasiparticle interference technique. Studying a conventional s-wave superconductor under applied magnetic field, we identified the relation between the odd-frequency superconducting pair correlations and different peaks in the change of the Fourier transformed local density of states directly measurable by STM/STS. Remarkably, we find a direct relationship between odd-frequency correlations and a bias asymmetry in the LDOS change. This method is applicable irrespective of the other symmetries of the Cooper pairs, due to it explicitly probing the oddness in frequency. We further validate this in the SM by showing the same LDOS change in a superconductor with spin-singlet p-wave odd-frequency pair correlations [66]. Similar QPI analysis can also be performed in other materials with bulk odd-frequency correlations, for example, multiband superconductors [27–30,32,33], Ising superconductors [67], non-Hermitian superconductors [26], heavy fermions [40], spin-3/2 systems with Bogoliubov Fermi surfaces [25], and also in heterostructures, to directly verify the existence of odd-frequency correlations. In fact, relevant QPI peaks, especially a bias asymmetry, have already been observed in heavy fermion [68] and iron-based multiband [54,55] superconductors; in the latter explained in terms of a sign-changing order parameter [54,55], but with no current consensus and with the possibility of odd frequency not yet explored. Our Letter thus provides a pathway in characterizing the pairing symmetries in novel superconductors.

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