Microwave Signature of Topological Andreev level Crossings in a Bismuth-based Josephson Junction

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Demonstrating the topological protection of Andreev states in Josephson junctions is an experimental challenge. In particular the telltale 4π periodicity expected for the current phase relation has remained elusive, because of fast parity breaking processes. It was predicted that low temperature ac susceptibility measurements could reveal the topological protection of quantum spin Hall edge states by probing their low energy Andreev spectrum at finite frequency. We have performed such a microwave probing of a phase-biased Josephson junction built around a bismuth nanowire, a predicted second order topological insulator, and which was previously shown to host one-dimensional ballistic edge states. We find absorption peaks at the Andreev level crossings, whose temperature and frequency dependencies point to protected topological crossings with an accuracy limited by the electronic temperature of our experiment.

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One of the striking properties of topological matter is protected metallic states at the interfaces between two insulators with different topological invariants. Those states have a unique dispersion relation with crossings of spin-momentum-locked Kramers partners at high symmetry points of the Brillouin zones, whose protection stems from the high spin-orbit interaction (SOI). Topological protection consequently allows for 1D ballistic transport (see, e.g., Refs. [1] for a review). When superconducting correlations are induced in a topological insulator (TI), particle-hole symmetry and fermion parity conservation enforce protected crossings of the Andreev eigenenergy levels at zero energy, which is often discussed in terms of Majorana modes [2–4], in contrast to avoided crossings of Andreev levels in topologically trivial materials. In this Letter, we demonstrate a protected crossing in a crystalline Bi nanowire connected to two S electrodes (a S-Bi-S junction) using a high frequency linear response experiment, confirming the second order topological character of bismuth [5].

Crystalline bismuth has been shown [6] to belong to the recently discovered family of higher order topological insulators. 3D second order topological insulators are insulating both in the bulk and at high symmetry surfaces, but possess metallic 1D channels at the hinges between surfaces with different topological indices [5]. The hinge states are helical and ballistic just like edge states in 2D topological insulators (2DTI). The recent prediction that bismuth is a second order topological material explains previous scanning tunneling microscopy experiments revealing 1D states along the edges of hexagonal pits in Bi (111) crystals [7], as well as Josephson current measurements on Bi nanowires [8,9] proximitized by superconducting contacts. Nevertheless, because of electron and hole pockets at bismuth's Fermi energy, the few hinge states are bound to coexist with many more nontopological bulk and surface states. Those nontopological states are sensitive to disorder, resulting in diffusive motion of charge carriers. There is therefore no visible signature of topological transport in a Bi nanowire connected to non superconducting contacts, since the conductance is dominated by the diffusive channels. The situation is fundamentally different when superconducting electrodes (S) connect the Bi nanowire. The supercurrent through the S-Bi-S junction then runs preferentially along the wire's narrow ballistic hinge states, as revealed by the magnetic field induced periodic interference pattern originating from the hinges' spatial separation [8,9], similar to S-2DTI-S junctions [10,11].

We recently demonstrated the ballisticity of the hinge states over micrometric distances via the measurement of a sawtooth-shaped current-phase relation (CPR) of a S-Bi-S junction [9]. Those experiments could not however prove the topological nature of the hinge states since the 4π periodicity expected of a protected Andreev level crossing was not observed. In fact, it is by now well understood that the 4π periodicity, a hallmark of topological Josephson junctions, cannot be observed in dc CPR measurements [12]. Two phenomena restore the 2π periodicity: the first is the quasiparticle poisoning that induces transitions between states of different parities at a given edge [12], the second is due to the coupling between hinge states of same parity on opposite sample edges [13]. By contrast, signatures of 4π periodicity were observed in ac Josephson effect measurements [14–16]. The interpretation of those experiments is however delicate since both nonadiabatic transitions in voltage-biased Josephson junctions [17] and topologically trivial Andreev states with energy close to zero [18] also lead to signatures of 4π periodicity.

An alternative proposal for the investigation of topologically protected zero-energy Andreev level crossings at π is to measure the ac linear orbital magnetic susceptibility of a phase-biased Josephson junction [19,20]. In contrast to dc CPR, the ac susceptibility not only probes the Andreev spectrum (in particular level crossings) but in addition reveals the relaxation timescales of the spectrum occupation (diagonal elements of the density matrix) and interlevel transitions (off-diagonal elements) [21,22]. Specifically, the adiabatic, low frequency response is just the (nondissipative) phase derivative of the CPR. At higher frequency, a nonadiabatic dissipative contribution appears. This contribution mostly consists of its diagonal component χ''_D, due to the relaxation of Andreev levels occupation. At low temperature, χ''_D is proportional to the current i carried by the highest occupied Andreev level and the phase derivative of its thermal occupation $\chi''_D(\varphi) \propto i\partial f/\partial\varphi = -i^2\partial f/\partial\epsilon$ (using that the current carried by the Andreev level of energy ϵ is $i = -\partial\epsilon/\partial\varphi$) [23].

As a result, a level crossing at zero energy translates into a peaked dissipative response χ''_D at $\varphi = \pi$, which diverges at zero temperature. This result is connected via the fluctuation-dissipation theorem to the prediction by Fu and Kane [2] that the phase-dependent thermal noise of the Josephson current in a topological junction should peak at π [24]. There is no such dissipation peak if the two levels anticross at π (with a small gap κ), since then the current is zero, and both the noise and ac dissipation are exponentially suppressed at temperature below κ. This dichotomy demonstrates the power of high frequency linear susceptibility and noise to probe the topological protection of edge or hinge states in a phase-biased topological insulator (see Fig. 1 and Supplemental Material [25]).

We have performed such ac phase-biased experiments by inserting an asymmetric superconducting quantum interference device (SQUID) built around a Bi nanowire into a multimode superconducting resonator (see Fig. 2). We find periodic absorption peaks, whose temperature and frequency dependencies point to topological crossings at π of the Andreev levels, to within less than 1/5th of the estimated electronic temperature $T_{el} = 100$ mK. This experiment also provides the characteristic relaxation time of Andreev levels occupation at π caused by fermion parity breaking due to quasiparticle poisoning.

The Bi nanowire-based asymmetric SQUID is connected to a $\lambda/4$ multimode resonator made of two parallel, one meter long, superconducting meander lines. The resonator is aligned to the asymmetric SQUID using standard e-beam lithography, followed by sputtering of 400 nm-thick Nb. We connect the resonator to the SQUID with focused-ion-beam-induced deposition of superconducting tungsten (see

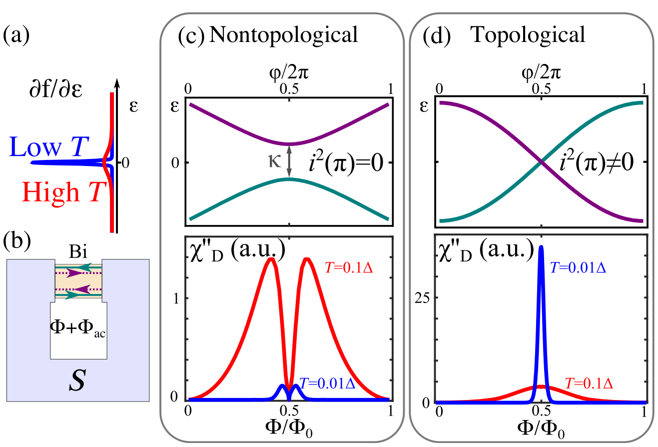

FIG. 1. Phase-dependent diagonal susceptibility as a signature of Andreev level crossings. (a) Derivative of the Fermi function entering in the expression of $\chi''_D \propto -i^2(\varphi)\partial f/\partial\epsilon$. (b) S-Bi ring threaded by a dc plus ac flux. (c) Topologically trivial Andreev spectrum displaying an avoided crossing at $\varphi = \pi$. This leads to a peak splitting of χ''_D at π since $i(\varphi = \pi) = 0$. (d) Topologically protected crossing at $\varphi = \pi$ with nonzero $i(\varphi = \pi)$, leading instead to a peaked χ''_D at π. The variations of χ''_D with phase are obtained from the tight binding computation of phase dependent Andreev bound states in an SNS junction on a hexagonal lattice with on site disorder: without SOI in (c) and with next-nearest-neighbor SOI in (d) at temperatures $T = 0.01\Delta$ (blue) and $T = 0.1\Delta$ (red), with Δ the superconducting gap (see Ref. [19] for details).

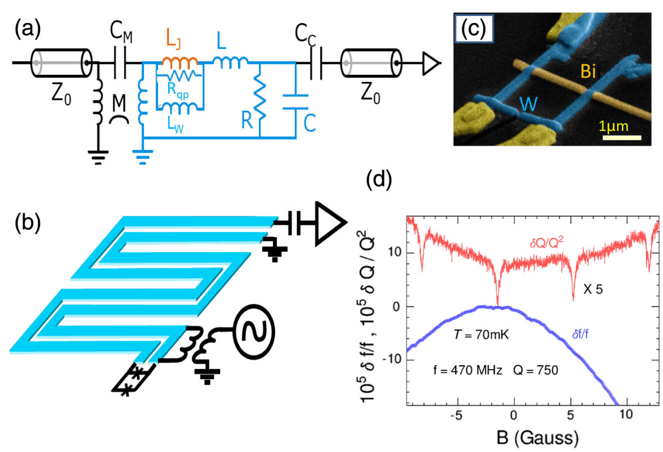

FIG. 2. (a) and (b) Principle of the experiment: the Bi nanowire is modeled by an inductance L_J in parallel with a resistance $R_r = 1/G_{qp}$ and a W wire symbolized by an inductance L_W. The SQUID is inserted in the resonator modeled by a R,L,C circuit measured in transmission with an inductive coupling to the microwave generator and a capacitive coupling to the cryogenic amplifier. (c) Scanning electronic micrograph of the Bi SQUID sample. (d) Field induced variations of the quality factor and frequency of the resonator's third eigenmode (average over 50 curves). We have arbitrarily fixed $\delta f = \delta Q = 0$ at $B = 0$. Note the sharp periodic absorption dips on $\delta Q(B)$ due to the Bi junction whereas the smooth parabolic shift $\delta f(B)$ is characteristic of the field dependent penetration depth of the resonator's Nb meander lines.

Fig. 2). These tungsten electrodes have a critical temperature $T_c \simeq 5$ K, which correspond to a superconducting gap $\Delta/k_B = 1.76T_c \simeq 9$ K. The resonator is measured in transmission, in a dilution refrigerator with base temperature 50 mK, using homodyne detection. The current's linear response $\delta I(t) = \delta I_\omega \exp -i\omega t$ to a small time-dependent flux $\delta\Phi_\omega \exp -i\omega t$ is characterized by the complex susceptibility $\chi(\omega, \varphi, T) = \delta I_\omega/\delta\Phi_\omega = i\omega Y(\omega, \varphi, T)$, where Y is the admittance of the NS ring, φ the superconducting phase difference, and T the temperature. As detailed in the Supplemental Material [25], the phase-dependence of the susceptibility's real and imaginary parts at the resonator's nth frequency f_n, $\chi'(\omega_n = 2\pi f_n, \varphi, T)$ and $\chi''(\omega_n, \varphi, T)$, is related to the variations of $\delta f_n(\Phi, T)$ and inverse quality factor $\delta(1/Q_n)(\Phi, T)$ induced by the dc magnetic flux Φ via:

$$\chi'(\omega_n, \varphi, T) = -\frac{L_R}{L_W^2}\frac{\delta f_n(\Phi, T)}{2f_n}$$

$$\chi''(\omega_n, \varphi, T) = \frac{L_R}{L_W^2}\delta\left(\frac{1}{Q_n}\right)(\Phi, T) \quad (1)$$

where $\varphi = -2\pi\Phi/\Phi_0$ with $\Phi_0 = h/2e$, $L_W \simeq 100$ pH the inductance of the W loop (including the W constriction) and $L_R \simeq 1$ μH the inductance of the resonator. We have previously conducted similar experiments on long SNS junctions in which the normal part N is a topologically trivial diffusive Au wire [22]. In those experiments, the susceptibility evolved from an adiabatic regime at low frequency, in which the susceptibility was exclusively nondissipative, given by the phase derivative of the Josephson current, to a dissipative regime at higher frequency, with minimal dissipation at π in agreement with theoretical predictions [26–29]. We report below a radically different behavior for the S-Bi-S junction: an exclusively dissipative susceptibility, peaked at π, that is compatible with topological protected Andreev states, see Fig. 1.

We measure the linear response for resonator eigenfrequencies ranging from 0.28 to 6.7 GHz. The response is periodic, with a period of 7G, corresponding to one flux quantum through the SQUID loop, as expected from the dc flux biasing we impose. The variations with field of the resonance frequency and quality factor are shown in Fig. 2(d) for the resonator's third eigenfrequency, $f_3 = 474$ MHz. The eigenfrequency shifts parabolically with field, as expected from the Nb resonator's kinetic inductance, but does not display periodic modulation. Thus at these frequencies, the response of the Bi-S ring is *not* the flux derivative of the dc Josephson current previously measured by SQUID interferometry [9], see Supplemental Material [25] By contrast, the quality factor displays below 0.5 K and for all eigenfrequencies, clear periodic dips that correspond to dissipation peaks in χ'' at odd multiples of $\Phi_0/2$ through the Bi-SQUID loop (i.e., a phase difference equal to π).

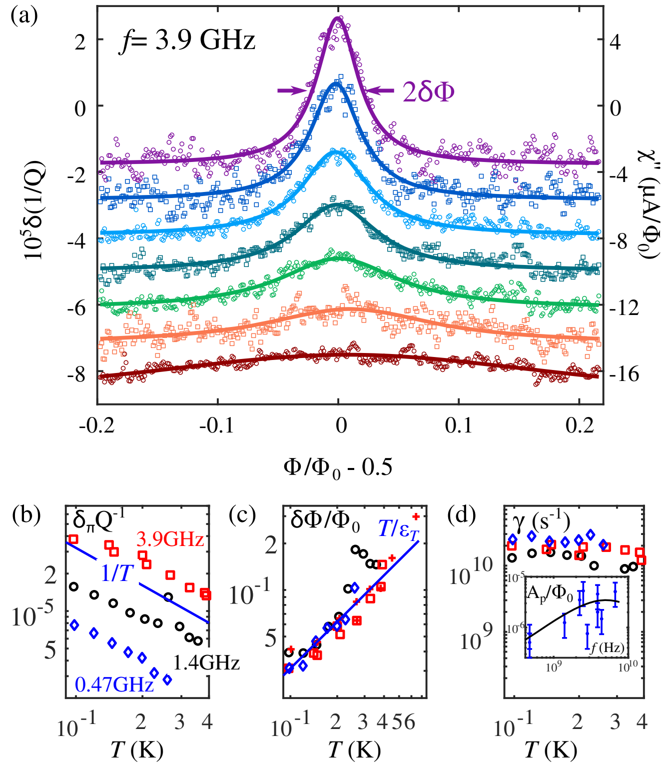

FIG. 3. (a) Temperature dependence of the dissipation peak, $\chi''(\varphi)$, around π at $f = 3.9$ GHz and $T = 0.1$, 0.15, 0.21, 0.27, 0.33, 0.39, 0.54 K from top to bottom (curves are shifted for clarity). Points are experimental data, solid line are fits of Eq. (2) to data taking ϵ_T as an adjustable parameter which is found to vary between 3 and 4 K. (b) and (c) Temperature dependence of the amplitude $\delta_\pi(\chi'') \propto \delta_\pi 1/Q$ and width $\delta\Phi/\Phi_0$ of the dissipation peaks around π, for different frequencies (diamonds 0.47 GHz, circles 1.4 GHz, squares 3.9 GHZ, crosses 4.5 GHz). The solid line in (c) corresponds to T/ϵ_T with $\epsilon_T = 3.5K$, the average extracted Thouless energy from our experimental data. (d) Main panel: relaxation rate γ deduced from the experiments of panel b. Inset: frequency dependence of the absorption peak area measured at 100 mK. A reasonable fit with Eq. (3) is obtained taking $\gamma = 3 \times 10^{10}$ s^{-1}, despite the dispersion in the data, mostly due to uncertainties in frequency-dependent calibration parameters.

The height of the dissipation peak $\delta_\pi(1/Q)$ at π varies as $1/T$, with no observable saturation down to 100 mK (see Fig. 3). It also increases linearly with frequency up to 4 GHz. Concomitantly, the peak width increases linearly with T and is independent of frequency. Thus the dissipation peak area is linear in frequency, with no temperature dependence. We show below that those results are consistent with the expected dissipative linear response of a two level Andreev spectrum with a nonavoided crossing at zero energy and $\varphi = \pi$.

Indeed, such an Andreev spectrum has the form $\epsilon(\varphi) = \pm\epsilon_T(\varphi/\pi - 1)$ near π, with ϵ_T the Thouless energy, estimated to $\epsilon_T \sim 4$ K from dc measurements. If we neglect the coupling between opposite edges of the wire, parity constraint and ballisticity impose that there is no coupling by the current operator between the levels and therefore no allowed interlevel transitions. The linear response's dissipative term χ'' must thus be restricted to its diagonal term, χ''_D, caused by the relaxation of thermal occupations of Andreev levels (see Supplemental Material [25]). It reads $\chi''_D = -2i_0^2[(\omega\gamma)/(\omega^2+\gamma^2)](\partial f/\partial\epsilon)$, which yields the following:

$$\chi''_D = i_0^2 \frac{\omega\gamma}{\omega^2+\gamma^2} \frac{1}{2T\cosh^2[\frac{\epsilon_T}{2T}(\varphi/\pi - 1)]}. \quad (2)$$

Here γ is the relaxation rate of the Andrev levels occupation and $i_0 = 2\epsilon_T/\Phi_0$ is the current carried by the Andreev states (in the long junction limit where ϵ_T is smaller than the superconducting gap). We note that this expression for the dissipative response is equivalent, via the fluctuation dissipation theorem, to the prediction of Fu and Kane for the noise power spectrum $S(\omega)$ through $S(\omega) = \hbar\coth(\hbar\omega/2k_BT)\chi''_D$. Figure 3(a) shows how well the simple expression Eq. (2) fits the experimental results, using an electronic temperature identical to the measured dilution refrigerator temperature down to 100 mK. The adjustable parameter is the Thouless energy ϵ_T which is found equal to 3.5 ± 0.5 K in agreement with its estimate from dc measurements [9]. We show in the Supplemental Material [25] that an avoided crossing at π due to a small coupling κ between levels would generate (because of the current going to zero) a split peak around π exponentially suppressed at temperatures below κ. Concomitantly, this coupling would also allow interlevel transitions, leading to an extra absorption peak at π whose width would be proportional to κ and independent of temperature. Since we see neither peak splitting nor temperature independent peak width, we conclude that there is a perfect level crossing to within our experimental energy resolution which is $T/5 = 0.4$ GHz at the lowest achieved electronic temperature of 100 mK (see Fig. S3). This result is a striking illustration of topological protection against disorder of Andreev level crossings originating from hinge states in these micron-long Bi nanowires. It is remarkable that these states can coexist with many diffusive surface states dominating the normal state transport ([9]).

We note that we have so far considered the contribution of only one pair of Andreev levels, i.e., a single hinge state, whereas two hinges carry the supercurrent (one at each acute angle) [9]. Those two hinges must be coupled at least at the wire ends where they are both connected to the superconductor [19]. Using a distance between edges $W_{\rm Bi} \simeq 200$ nm and a superconducting coherence length $\xi_W \simeq 20$ nm, we estimate this coupling to be $\kappa = \epsilon_T \exp(-W_{\rm Bi}/\xi_W) \simeq 0.2$ mK, which is about 500 times smaller than the experimental temperature. This justifies our approximation of uncoupled hinge states. In addition, our previous experiments [9] indicate that one edge carries a current four times larger than the other and therefore yields the main contribution to χ''_D by a factor 16.

We have shown that the peaked χ'' signals a nonavoided level crossing at π and a thermal occupation of the levels. We now discuss the rate at which the relaxation to thermal equilibrium occurs. Since spin-orbit coupling prevents direct transitions between spin-locked Andreev levels within one hinge, and since the coupling between the hinges is negligible, the most effective relaxation mechanisms must be due to quasiparticle poisoning. Such quasiparticles originate either from the superconducting W [30] or from nontopological (surface or bulk) states in the bismuth wire. We extract the rate of relaxation to equilibrium from the dissipation peak area $A_p(\omega) = \delta_\pi\chi''\delta\Phi$,

$$A_p(\omega) = i_0\omega\gamma/(\omega^2+\gamma^2), \quad (3)$$

using $i_0 = 400$ nA determined in Ref. [9]. This yields a relaxation rate $\gamma \simeq 2 \pm 1 \times 10^{10}$ s^{-1}, that is temperature independent up to 0.5 K, see Fig. 3(d). Fu and Kane [2] suggested that $\gamma(\pi, T)$ is the exchange rate between the zero energy Andreev states $\Psi_A(\pi)$ of the W-Bi-W junction and quasiparticles at finite energy. In a hard gap superconductor, this rate is exponentially suppressed at temperatures below the gap [31,32]. Our observation that γ is independent of temperature below 0.5 K points to the presence of low energy quasiparticles in the circuit. Following [31], γ is deduced from the Fermi golden rule:

$$\gamma = 2\pi^2 \int n_{\rm qp}(\epsilon)\left(1 - f\left(\frac{\epsilon}{k_BT_{\rm el}}\right)\right) f_{\rm BE}\left(\frac{\epsilon}{k_BT_{\rm env}}\right) \times |\langle\Psi_A|\mathbf{I}|\phi_{\rm qp}(\epsilon)\rangle|^2 \Re\left[Z\left(\frac{\epsilon}{\hbar}\right)\right]\frac{d\epsilon}{\epsilon} \quad (4)$$

Absorption of a photon at energy ϵ from the electromagnetic environment of the Bi junction gives rise to a quasiparticle emission at the same energy with a probability $P(\epsilon)$ proportional to $\Re[Z(\epsilon/\hbar)]$, the real part of the impedance in parallel with the Bi wire, f and $f_{\rm BE}$ are the Fermi and Bose-Einstein distribution functions, respectively taken at the electronic ($T_{\rm el}$) and environment ($T_{\rm env}$) temperatures, $\langle\Psi_A|\mathbf{I}|\phi_{\rm qp}(\epsilon)\rangle$ is the matrix element of the current operator between the Andreev and quasiparticle states. In the Supplemental Material [25], we estimate γ from the electromagnetic environment of the Bi nanowire. This environment is determined by the quasiparticle resistance in parallel with the kinetic inductance of the Bi wire, and the impedance of the resonator (coupled to the RF measurement circuit). A value of $\gamma \simeq 10^{10}$ s^{-1}, close to our experimental findings, is obtained if we take a photon temperature $T_{\rm env}$ of the order of 2 K. This high effective temperature compared to $T_{\rm el}$ could be due to the resonator's broadband coupling to the cryogenic microwave amplifier. We thus attribute the high relaxation rate γ in our experiment to a sizable density of unpaired quasiparticles in the SQUID [30]. These poisoning processes could in principle be considerably suppressed by using a hard gap superconductor to contact the Bi nanowire and working with a single mode resonator with a narrow bandwidth [33].

There is one apparent inconsistency, however. The high relaxation rate means that $\omega/\gamma \leq 1$ for most eigenfrequencies we probe, and therefore the response regime should be quasiadiabatic. This implies that $\chi'(\varphi)$ should be proportional to the derivative of the CPR that we measured on this very same sample in the previously reported experiment [9]. The fact that we detect no $\chi'(\varphi)$ may indicate that the even and odd parity levels are equally populated around π because of the fast relaxation within one hinge. Since the two parity levels carry opposite current, this would cancel $\chi'(\varphi)$ but not $\chi''_D(\varphi)$ proportional to i^2. In contrast, the CPR experiment [9] was conducted at low frequency (10^4 to 10^5 Hz) compared to the interhinge rate $\kappa/h \simeq 5$ MHz, so that during the CPR measurement both edges can be explored, restoring the CPR of a long ballistic nontopological 1D wire [2,13].

We have therefore obtained a consistent picture of the phase-dependent, high-frequency linear response of a Bi-based Josephson junction whose dissipation peaks at π reveal protected Andreev level crossings and thus the topological character of Bi. The short relaxation time we find (0.1 ns) is most likely due to subgap quasiparticle poisoning processes and to the coupling to an insufficiently thermalized electromagnetic environment. The comparison between dc and ac experiments suggests a greater, μs long, interhinge scattering time. These results call for future measurements in the MHz range, to explore fermion parity exchange processes between opposite hinges [13]. Much higher frequency (of the order of the Thouless energy) should also be interesting, revealing parity conserving transitions of the long junction Andreev spectrum discussed in Refs. [34,35,36].

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