Pauli-Limited Multiband Superconductivity in KFe$_2$As$_2$

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The upper critical field $H_{c2}(T)$ of KFe$_2$As$_2$ has been studied via low-temperature thermal expansion and magnetostriction measurements. The observation of quantum oscillations of the sample length associated with orbits in its five hole-like Fermi surfaces suggest that the interaction of these bands strongly influences the superconductivity in KFe$_2$As$_2$ and manifests in the multigap shape of our thermal-expansion measurements. We present compelling evidence for Pauli-limiting effects dominating $H_{c2}(T)$ for $H \parallel \text{a}$, as revealed by a crossover from second- to first-order phase transitions to the superconducting state in the magnetostriction measurements down to 50 mK. Corresponding features were absent for $H \parallel \text{c}$. To our knowledge, these results constitute the first confirmation of Pauli limiting of the $H_{c2}(T)$ of a multiband superconductor. These results are supported by modeling Pauli limits for single-band and multiband cases.

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The iron-based multiband SCs present a unique opportunity to study these matters in detail. Here, we present measurements on KFe$_2$As$_2$ single crystals, which give evidence for a Pauli-limited multiband SC. KFe$_2$As$_2$ crystallizes in a tetragonal ThCr$_2$Si$_2$-type structure (space group I4/mmm). It is the end-member of the Ba$_{1-x}$K$_x$Fe$_2$As$_2$ series, in which the superconducting state reaches a maximum $T_c$ of 38 K at $x \sim 0.4$ [9]. Due to the proximity of these compounds to antiferromagnetic order, their pairing mechanism is believed to arise from magnetic fluctuations, as it is discussed for cuprate and heavy-fermion SCs [10]. For KFe$_2$As$_2$, evidence for multigap nodal s-wave superconductivity has indeed been found in nuclear quadrupole resonance [11] and angle-resolved photoemission spectroscopy (ARPES) [12], while recent thermal-conductivity measurements suggest d-wave pairing [13].

Compared to optimally doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$, the low superconducting transition temperature $T_c \sim 3.4$ K of KFe$_2$As$_2$ allows us to explore its entire $H$-$T$ phase diagram. We performed thermal-expansion and magnetostriction measurements in a temperature range between 50 mK and 4 K and in magnetic fields up to 14 T applied parallel and perpendicular to the c-axis of the crystals. Our experiments constitute an extension of the measurements performed above 2 K by Burger et al. [14], in which initial evidence of strong Pauli-limiting effects was presented. The experiments were carried out in a home-built capacitive dilatometer. The linear thermal-
expansion and magnetostriction coefficients are defined as
\[ \alpha_i = L_i^{-1} \partial L_i / \partial T \]
and \[ \lambda_i = L_i^{-1} \partial L_i / \partial (\mu_0 H) \], respectively, where \( L_i \) is the length of the sample along the \( i = a, c \)-axis. As \( \alpha_i \) is related to the uniaxial pressure dependence of the entropy \( S \) via Maxwell relations, we can use \( \alpha_i \) to search for nearby pressure-induced instabilities.

Single crystals of KFeAs were grown in a K-Fe-As melt rich in K and As to reduce the amount of magnetic impurities [13]. The residual resistivity ratio (RRR) of the samples amounts to \( \sim 1000 \) [15]. As flux-grown iron arsenides tend to form foliated stacks with embedded flux, the observation of quantum oscillations (QOs) for both field directions in our magnetostriction measurements represents a particularly reliable quality probe.

The mean-free-paths (mfp) determined from the Dingle temperature of the QOs amount to \( \ell = 52 \pm 3 \) nm along the \( a \)- and \( c \)-axis, respectively. With the coherence lengths of \( \xi_a \sim 15 \) nm and \( \xi_c \sim 3 \) nm [17], the ratio \( \ell / \xi_c \sim 15 \) confirms that the samples are in the superconducting clean limit. The extracted high effective masses are consistent with the enhanced Sommerfeld coefficient. Furthermore, the FS cross-sectional areas inferred from our data are in agreement with the reported electronic structure in which the contribution of each band to the FS differs in its dimensionality [15]. Further details about the QOs of the magnetostriction will be given in a separate publication.

The linear thermal-expansion coefficients \( \alpha/T \) of KFeAs are plotted in Fig.1(a) for \( H || a \). For \( H = 0 \), the SC transition has a step-like form, with no appreciable difference between cooling and heating curves. Besides the steps at \( T_c \), the data for \( H = 0 \) show additional broad maxima at \( \sim 0.5 \) K, displayed in more detail in Fig.1(b) for both \( \alpha_a/T \) and \( \alpha_c/T \). These features directly manifest the superband nature of superconductivity in KFeAs. A shoulder of \( C/T \) vs. \( T \) has previously been observed in KFeAs [14], similar to that of the well-known multiband SC MgB\(_2\), in which the observed feature is caused by the opening of a low-energy superconducting gap on one of the weakly coupled bands [19]. Even though \( \alpha_i/T \) and \( C/T \) are interrelated via the Gruneisen parameter, the maxima in \( \alpha_i/T \) are much more pronounced than the ones observed in \( C/T \).

For \( H > 0 \) [Fig.1(a)], the system enters an irreversible regime, possibly due to vortex pinning effects. As \( H \) is increased and \( T_c \) is suppressed, a clear increase of \( \alpha_c/T \) emerges at \( \mu_0 H = 4 \) T (\( T_c \sim 1.7 \) K) and continues to develop to a peak-like transition at higher fields. The increase of \( \alpha(T, H)/T \) for large fields resembles a crossover from a SO to a FO phase transition, expected for a system presenting strong Pauli-limiting effects. Evidence for Pauli-limiting effects in KFeAs has been reported in earlier measurements of \( H_{c2}(T) \) [12] and magnetization [14]. For \( H > H_{c2} \) (5 T curves in Fig.1), \( \alpha_i/T \) do not show any strong divergence down to 100 mK that could be related to quantum critical behavior, ruling out the presence of nearby pressure-induced instabilities.

The SO–FO crossover becomes strikingly visible in the magnetostriction data displayed in Fig.2. For \( H || a \), a discontinuous variation of the sample length develops at \( H_{c2} \) as the field is swept at low-\( T \) [Fig.2(a)]. Clearly, this discontinuity is not present for \( H || c \) [Fig.2(b)]. The first-order-like length discontinuities observed at low temperatures for \( H || a \) translate into the very pronounced peaks of the length derivatives of the length derivatives \( \lambda_i(H, T) \) displayed in Fig.2(c). At 50 mK, the maximum value of \( \lambda_c(H) \) is almost 20 times larger than the transition step at 3 K. The values of \( \lambda_i^{\text{max}} \) are plotted in the projected \( \lambda_c-H \) plane, from which it is possible to define a SO–FO crossover temperature \( T_0 \sim 1.5 \) K [see also Fig.3(b)]. Magnetic-field hysteresis is also observed at low temperatures, consistent with the FO character of the transition, and appears to be suppressed above 500 mK [15]. We have ruled out, on the basis of a detailed examination of the hysteretic behavior [15], the possibility that the FO transition could arise from the onset of the irreversible regime of the vortex lattice, which appears at magnetic fields slightly smaller than \( H_{c2}^{ab} \) at 50 mK and persists even where SO superconducting transitions are observed, for example, at 2.5 K for \( H || a \) and 50 mK for \( H || c \).

The \( H-T \) phase diagram derived from our \( \alpha_i \) and \( \lambda_i \) measurements is presented in Fig.3(a). While \( H_{c2} \) in-
FIG. 2: Changes in sample length $\Delta L = L - L_0$ measured along the $c$-axis of the crystal versus magnetic field (a) $H \parallel a$ and (b) $H \parallel c$ at different temperatures ($L_0 = 500 \mu m$). (c) Magnetostriiction $\lambda_c$ vs. $H \parallel a$ for $0.05 K \leq T \leq 3 K$. $\lambda_c$ maxima are plotted in the projected $\lambda_c - T$ plane (blue circles), from which a tricritical temperature $T_0 \sim 1.5 K$ can be extracted.

The calculated single-band $H_{c2}(T)$ curves are displayed in Figs. 3(a). They adjust well the experiment for both field directions. For $H \parallel ab$, the calculations give a Maki parameter $\alpha_M = 3.8$, consistent with the observation of a FO phase transition. On the other hand, the calculations predict $T_{0}^{PB} \sim 1 K$, significantly smaller than $T_{0}^{FP} \sim 1.5 K$ determined from $\lambda_c^{max}$. This is remarkable, as the calculated value should constitute an upper limit: $T_0$ is determined by $\alpha_M$, i.e. the balance between Pauli and orbital pair-breaking, $T_0$ is hardly changed by antiferromagnetic (AFM) fluctuations, nodes in the gap function, or strong-coupling effects [22]. Disorder, on the other hand, suppresses the Pauli pair-breaking effects and reduces $T_0$ [17]. The discrepancy between a rather high $T_0^{FP}$ and the single-band $\alpha_M$ is further illustrated by a comparison with other Pauli-limited SCs. The organic SC $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, for example, has a comparable temperature $T_0 = T_0^{FP}$, determined by the peak height of $C/T$, but a much higher $\alpha_M = 8$ [8]. In a clean-limit SC, the orbital pair-breaking effects are determined by the Fermi velocity of the shielding currents which can be extracted from the slope $H_{c2}$ at $T_c$. The deviation of $\alpha$ decreases monotonically with decreasing $T$, $H_{c2}^{ab}$ flattens out below $1.5 K$, a sign of strong Pauli-limiting effects. Moreover, $H_{c2}^{ab}(0) = 4.8 T$, which is much smaller than the clean-limit orbital field $0.73 T_c dH_{c2}/dT |_{T_c} \sim 15.4 T$. The crossover to a discontinuous phase transition at $T_0$ can only be observed for $H \parallel ab$ (Figs. 1 and 2), the field direction for which shielding currents are minimal. This suggests that the driving force for Pauli limitation in KFe$_2$As$_2$ is the quasi-two-dimensional electronic structure, in contrast to CeCoIn$_5$ where FO transitions appear for both field directions [3]. Despite the clear indications of Pauli limitation in $H_{c2}^{ab}$, our data does not show signatures of a possible FFLO phase at high fields such as a double transition or an upturn of $H_{c2}(T)$ towards low $T$ observed in $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ [3]. We cannot rule out the possibility of a slight misalignment of the magnetic field with the sample inhibiting the formation of the FFLO phase [21]. Remarkably, $H_{c2}^{ab}$ and $H_{c2}^{c}$ of KFe$_2$As$_2$ exhibit a $T$-dependent anisotropy factor $\Gamma = H_{c2}^{ab}/H_{c2}^{c}$ [Fig. 3(a)], contrary to the constant anisotropy expected from Ginzburg-Landau theory. This unusual anisotropy has also been reported for LiFeAs and Fe(Se,Te) [22–24], and has been attributed to Pauli limiting and/or multiband effects.

To interpret theoretically our results, we model $H_{c2}(T)$ using first a single-band formalism. We considered the solutions to the linearized Gor’kov equations developed by Werthamer, Helfand and Hohenberg (WHH) for a uniaxial, clean-limit SC, following the approach recently presented by Gurevich [8, 25, 26]. This model takes into account orbital and Zeeman pair-breaking effects, as well as the formation of an FFLO state below a tricritical temperature $T_0$ when its modulation wavelength $\lambda_0$ is shorter than the mfp $\ell$. Apart from the $H_{c2}(T)$ curve, the model yields the Fermi velocities and the Pauli susceptibility $\chi_N = (1/2)\mu_B^2 N(E_F) / \gamma$ by obtaining the gromagnetic factor $g$. It also determines the FFLO phase boundaries below $T_0$ and the modulation vector $Q \propto \lambda_0^{-1}$, although these values should be taken with caution as they are sensitive to details of the electronic band structure and disorder which are not considered in the model. The values of $\nu_F$ were always kept within the range of the values deduced from our QOs (see [17] for a summary of the parameters used in the calculations).

The values of $\nu_F$ were always kept within the range of the values deduced from our QOs (see [17] for a summary of the parameters used in the calculations).
\( H_{c2}(0) \) from \( H_{c2}^{orb} \), on the other hand, represents a measure of Pauli-limiting effects. As \( T_c, H_{c2}, \) and \( H_{c2}(0) \) are the only free parameters in the model, the irreconcilable difference between \( T_0^{exp} \) and \( T_0^{1B} \) suggests the impossibility of describing KFe\(_2\)As\(_2\) with a single-band model.

The failure of the single-band model in explaining the \( H-T \) phase diagram of KFe\(_2\)As\(_2\) becomes even more evident for \( H \parallel c \), where calculations result in the unphysical value \( g_c \approx 0 \), although values of \( g < 2 \) might be possible in the presence of the Jaccarino-Peter effect \[28\]. Since AFM fluctuations are indeed present in KFe\(_2\)As\(_2\) with the magnetic easy plane perpendicular to the \( c \)-axis, a reduced \( g \)-factor should be visible for \( H \parallel ab \) and not for \( H \parallel c \). Furthermore, the extreme magnetic anisotropy indicated by \( g_{ab}/g_c \rightarrow \infty \) as a result of a single-band model clearly contradicts magnetization and Knight-shift measurements which unambiguously reveal a nearly isotropic susceptibility, with \( \chi_{ab}/\chi_c \approx 1.2-1.5 \) \[16, 29, 30\].

If more than one band contributed to the FS, the slope \( H_{c2}' \) would be proportional to a superposition of Fermi velocities, \( H_{c2}' \propto (\sum c_n v_{F,n})^{-1} \). The coefficients \( c_n \) are functions of the superconducting coupling constants. Since bands differing in shape and dimensionality could essentially yield very different values of \( v_{F,n} \), it is very well conceivable that one band could be Pauli limited while the others remained orbitally limited. In this case, a SO–FO crossover would occur below a high value of \( T_0 \) even for a relatively small slope \( H_{c2}' \). The FFLO state, on the other hand, could be damped by the bands with dominating orbital pair-breaking. The electronic structure of KFe\(_2\)As\(_2\) inferred from our QOs and recent ARPES measurements does indeed reveal bands of different characteristics \[12, 18\]. The five bands that cross the Fermi energy and hence contribute to the FS, however, cannot be considered in the model due to the complexity of the calculations. In order to capture the basic physical description, we restrict the calculations to a two-band model, for which four coupling constants enter as additional parameters. We therefore adjust the data within two extreme scenarios: one dominated by inter-band coupling, and the other by intra-band coupling, as proposed for Fe-based SCs and for MgB\(_2\), respectively. The latter scenario is supported by our thermal-expansion measurements which show similarities with this material.

The results of the two-band calculations are presented in Fig. 3(b), showing that this model moves \( T_0^{1B} \) to higher temperatures compatible with the experiment, while keeping \( H_{c2}' \sim -6 \) T/K. With these parameters, \( H_{c2}(T) \) is practically independent of the coupling constants. The higher \( T_0^{1B} \) leads to a more extended stability range of the FFLO state compared to the single-band calculations. The band which is less affected by Pauli limiting inhibits, however, the formation of an FFLO state (smaller \( Q \)) in the multi-band case, resulting in a larger value of \( \lambda_Q \) which exceeds \( \ell_{ab} \) \[13\]. The effect of suppression of the FFLO phase by non-Pauli-limited bands is expected to be stronger if all five bands involved in the electronic structure of KFe\(_2\)As\(_2\) are considered in the model.

Multiband superconductivity is ubiquitous in Fe-based superconductors. In Ba\(_{1-x}\)K\(_x\)Fe\(_2\)As\(_2\), increasing the K content lowers the dimensionality of the electronic structure and gives rise to strong correlations. These conditions favor Pauli pair-breaking effects in KFe\(_2\)As\(_2\), where we found compelling evidence for Pauli-limited multiband superconductivity. In more general terms, our experiments have shown the complex interplay of pair breaking and multiband effects, which have to be taken into account in models of multiband superconductivity in iron-based superconductors \[31\].

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**Figure 3:** (a) Left axis: \( H_{c2}^{1B} \) (open symbols) and \( H_{c2}^{cc} \) (closed symbols) vs. \( T \) for KFe\(_2\)As\(_2\), determined from \( \alpha(T) \) (triangles) and \( \lambda(T) \) (squares). Solid green lines correspond to single-band calculations, with the vertical arrow indicating the position of the corresponding tricritical-point temperature \( T_0^{1B} \), below which the FFLO phase is predicted to form. Right axis: the \( H_{c2} \) anisotropy factor \( \Gamma \) vs. \( T \) (blue circles). (b) Two-band calculations for \( H \parallel ab \). The limiting cases of dominant inter (intra) band coupling are indicated by solid (dashed) lines. Arrows indicate the position of the tricritical-point temperature from \( \lambda_{c2}^{max} (T_0^{1B}) \) and from the calculations \( (T_0^{1B}) \). \( Q \)-vector amplitudes vs. \( T \) obtained from single- and two-band calculations are also displayed. In (a) and (b), the calculated upper lines below \( T_0 \) represent the onset of the FFLO state, while the lower line corresponds to the onset of the homogeneous phase with \( Q = 0 \).
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Pauli-Limited Multiband Superconductivity in KFe$_2$As$_2$

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I. SUPPLEMENTAL MATERIAL

A. SAMPLE PREPARATION

Single crystals of KFe$_2$As$_2$ were grown from self flux. With FeAs flux, only samples of minor quality and small size were obtained. Fluxes rich in As and K yield high quality single-crystals of large size ($\sim 5 \times 5 \text{mm}^2$). The appropriate amounts of K, As, and pre-reacted FeAs or FeAs$_2$ were combined in an Al$_2$O$_3$ crucible and sealed in a steel cylinder under 1 bar of argon. Following the heating of the cylinder to $920–1000^\circ\text{C}$, the crystal growth was started by slowly cooling the furnace to $730–850^\circ\text{C}$ at rates of $0.3–0.49^\circ\text{C}/\text{hour}$. Once the lower temperature was reached, the cylinder was tilted to separate the crystals from the remaining liquid flux, followed by a slow cool-down to room temperature. After opening the containers, it was possible to collect the free-standing single crystals with tweezers, while an etching treatment with ethanol was necessary to separate the remaining crystals trapped in the solid flux. Samples with typical residual-resistivity ratios $\text{RRR} = \rho(300\text{K})/\rho(4\text{K}) \sim 1000$ were obtained [1].

B. QUANTUM OSCILLATIONS IN MAGNETOSTRICTION

Fig. S1 displays the quantum oscillations (QOs) observed in magnetostriuction measurements of the sample length along the $a$–axis of the sample and for applied magnetic fields $H \parallel c$. Peaks are labeled following previous de Haas–van Alphen (dHvA) measurements [2]. Details of the analysis of the QOs data (effective masses, mean free paths, etc.) will be presented in a separate manuscript.

![High-field magnetostriction $\lambda_a$ of KFe$_2$As$_2$ for magnetic fields $H$ applied along the $c$–axis at several temperatures.](a)

![Fourier-transform spectra of the quantum oscillations displayed in (a).](b)

FIG. S1: (a) High-field magnetostriction $\lambda_a$ of KFe$_2$As$_2$ for magnetic fields $H$ applied along the $c$–axis at several temperatures. (b) Fourier-transform spectra of the quantum oscillations displayed in (a).

C. HYSTERESIS IN MAGNETOSTRICTION

The data displayed in Fig. S2(a) shows a clear shift of the rising and trailing edges between the 100mK magnetostriction curves obtained upon increasing and decreasing magnetic field ($H \parallel a$), which we attribute to hysteresis effects associated with the first-order phase transition. This hysteresis appears to be suppressed above 500mK [Fig. S2(b)] except for small changes in the height of the maxima. $H_{c2\uparrow}$ obtained upon increasing $H$ at a rate $\mu_0\dot{H} = 0.04\text{T/min}$ is shifted with respect to $H_{c2\downarrow}$ measured while decreasing $H$ at $\mu_0\dot{H} = −0.04\text{T/min}$. The shift
\( \mu_0 \Delta H_{c2} = \mu_0 |H_{c2+} - H_{c2-}| \approx 0.02\, \text{T} \) corresponds to a time shift \( \Delta t \approx 30\, \text{seconds} \) for \( \mu_0 \dot{H} = 0.04\, \text{T/min} \), which cannot be attributed to a measurement delay since it is much higher than the delay of our apparatus, reflected in the almost identical curves measured with different rates, \( \mu_0 \dot{H} = -0.04\, \text{T/min} \) and \( \mu_0 \dot{H} = -0.02\, \text{T/min} \).

\[ \frac{dH}{dt} = \frac{\Delta H}{\Delta t} \]

**FIG. S2**: Magnetostriction \( \lambda_c \) vs. field at (a) 100 mK and (b) 500 mK.

The hysteresis is also observed directly in the length change, as shown in more detail in Fig. S3(a) for \( H \parallel a \) and \( T = 50\, \text{mK} \). On the other hand, the curves measured in decreasing (blue) and increasing (red) magnetic field separate notoriously right below the superconducting transition at \( H_{c2}^{ab}(50\, \text{mK}) \) (green arrow), which might indicate the entrance into the irreversible vortex state. The splitting of the two curves persists in the measurements taken at \( T = 2.5\, \text{K} \) for \( H \parallel a \) [Fig. S3(b)] and at \( T = 50\, \text{mK} \) for \( H \parallel c \) [Fig. S3(c)], where the superconducting transition is always of second order.

**FIG. S3**: \( c \)-axis sample length \( L_c \) (left axis) and magnetostriction \( \lambda_c \) (right axis) versus applied magnetic field \( H \). (a) \( H \parallel a \), \( T = 50\, \text{mK} \); (b) \( H \parallel a \), \( T = 2.5\, \text{K} \); (c) \( H \parallel c \), \( T = 50\, \text{mK} \). The green arrows indicate the value of magnetic field below which the measurements taken during decreasing (blue) and increasing (red) magnetic field separate.

**D. Upper Critical Field Calculations**

The calculations of \( H_{c2}(T) \) were performed using the model developed by Werthamer, Helfland and Hohenberg (WHH) for a uniaxial, clean-limit SC \([3, 4]\), following the approach recently presented by A. Gurevich \([5]\). For the single-band (1B) calculations the equation

\[
\ln t + U(t, b, q) = 0
\]

has to be solved, which involves the calculation of \( U(t, b, q) \)

\[
U(t, b, q) = 2e^2 \text{Re} \sum_{n=0}^{\infty} \int_{q}^{\infty} du \, e^{iu^2} \left\{ \frac{u}{n+1/2} - \frac{t}{\sqrt{b}} \tan^{-1} \left[ \frac{u \sqrt{b}}{t(n+1/2) + i \alpha G b} \right] \right\}
\]
where \( t = T/T_c \), and \( b, q \) and \( \alpha_G \) are the reduced upper critical field, reduced magnitude of the \( Q \)-vector corresponding to the FFLO phase, and reduced Maki parameter, respectively. In this model, an FFLO phase appears if \( \alpha_G \geq 1 \), and it is related to the actual Maki parameter via \( \alpha_G \approx \alpha_M / 1.845 \). The band anisotropy can be described in terms of the ratio of the effective masses or the ratio of the Fermi velocities:

\[
\epsilon = \frac{m_\perp}{m_\parallel} = \left( \frac{v_\parallel}{v_\perp} \right)^2 \tag{3}
\]

where \( \parallel \) and \( \perp \) denote the directions parallel and perpendicular to the direction of the applied magnetic field:

\[
\text{for } H \parallel c : \quad v_\parallel = v_c, \quad v_\perp = v_{ab} \quad \rightarrow \quad \epsilon_c = \left( \frac{v_c}{v_{ab}} \right)^2 \tag{4}
\]

\[
\text{for } H \parallel ab : \quad v_\parallel = v_{ab}, \quad v_\perp = \sqrt{v_{ab} v_c} \quad \rightarrow \quad \epsilon_{ab} = \frac{v_{ab}}{v_c} \tag{5}
\]

The shielding currents are determined by the component of the velocity perpendicular to the field, i.e., \( \frac{dH_c}{dT} \bigg|_{T_c} \propto \frac{1}{v_\perp} \).

For a two-band (2B) calculation, the equation to solve is

\[
a_1(\ln t + U_1) + a_2(\ln t + U_2) + (\ln t + U_1)(\ln t + U_2) = 0 \tag{6}
\]

where \( U_1 \) corresponds to Eq. 2 and \( U_2 \) is defined as:

\[
U_2(t, b, q) = 2e^2 s \Re \sum_{n=0}^\infty \int_{q \sqrt{s}}^\infty du \, u^2 \left\{ \frac{u}{n + 1/2} - \frac{t}{\sqrt{bn}} \tan^{-1} \left[ \frac{u \sqrt{bn}}{t(n + 1/2) + i \alpha_G b} \right] \right\} \tag{7}
\]

with the inter-band parameters \( s \) and \( \eta \) defined as:

\[
s = \frac{\epsilon_2}{\epsilon_1}, \quad \eta = \left( \frac{v_{ab,2}}{v_{ab,1}} \right)^2 \tag{8}
\]

The parameters \( a_1 \) and \( a_2 \) in Eq. 4 are defined as:

\[
a_1 = \frac{\lambda_0 + \lambda_-}{2w}, \quad a_2 = \frac{\lambda_0 - \lambda_-}{2w} \tag{9}
\]

\[
\lambda_\pm = \lambda_{11} \pm \lambda_{22}, \quad \lambda_0 = \sqrt{\lambda_2^2 + 4\lambda_{12}\lambda_{21}}, \quad w = \lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21}. \tag{10}
\]

and determine the level of intra- and inter-band coupling of the calculations:

- \( \lambda_{11}\lambda_{22} << \lambda_{12}\lambda_{21} \rightarrow \) inter band coupling limit (2B-inter)
- \( \lambda_{11}\lambda_{22} >> \lambda_{12}\lambda_{21} \rightarrow \) intra band coupling limit (2B-intra)

The following tables summarize the parameters used in the single- and two-band calculations, where \( g \) corresponds to the gyromagnetic factor, and \( \lambda_Q^{\text{min}} \) is proportional to the inverse of the maximum value of \( Q \) displayed in Fig. S1(a).

**TABLE S1: Single-band calculation parameters.**

| \( H \)-field direction | \( T_c \) (K) | \( g \) | \( v_{ab} \) (m/s) | \( v_c \) (m/s) | \( \frac{dH_c}{dT} \bigg|_{T_c} \) (T/K) | \( H_{c2}^{\text{orb}} \) (T) | \( H_{c2}^P \) (T) | \( \alpha_M \) | \( T_0^{1B} \) (K) | \( t_0 = \frac{T_0^{1B}}{T_c} \) | \( \lambda_Q^{\text{min}} \) (nm) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| ab | 3.4 | 2.24 | 42500 | 4100 | -6.2 | 15.33 | 5.65 | 3.84 | 1.01 | 0.295 | 52 |
| c | 0 | 42500 | 4100 | -0.6 | 1.48 | \( \infty \) | 0 | - | - | - | - |
TABLE S2: Two-band calculation parameters, for strong inter band coupling scenario.

| H-field direction | Coupling parameters | $T_c$ (K) | $g$ | $\frac{dH_{c2}}{dT_c}$ (T/K) | $T_{c2B}$ (K) | $t_0 = \frac{\gamma_{c2B}^{2B}}{\gamma_{c2}^0}$ (nm) |
|-------------------|---------------------|-----------|-----|-----------------------------|---------------|-----------------------------------------------|
| $\lambda_{11}$    | $\lambda_{22}$      | $\lambda_{12}$ | $\lambda_{21}$ | 3.4 | 2.23 | -5.99 | 1.43 | 0.42 | 495 |
| $\lambda_{22}$    | 0                   | 0         | 0.5 | 0.5 | 2     | -0.332 | - | - | - |

| H-field direction | Band 1                  | Band 2                  |
|-------------------|-------------------------|-------------------------|
| $v_{ab}$ (m/s)    | $v_c$ (m/s) | $H_{c2B}^{2B}$ (T) | $H_{c2}^{c2}$ (T) | $\alpha_M$ | $v_{ab}$ (m/s) | $v_c$ (m/s) | $H_{c2B}^{2B}$ (T) | $H_{c2}^{c2}$ (T) | $\alpha_M$ |
| $ab$              | 28000                  | 11700                   | 8.15 | 6.31 | 2.23 | 0.82 | 1.38 | 0.41 | 244 |
| $c$               | 28000                  | 11700                   | 3.40 | 6.31 | 2.23 | 0.82 | 1.38 | 0.41 | 244 |

TABLE S3: Two-band calculation parameters, for strong intra band coupling scenario.

| H-field direction | Coupling parameters | $T_c$ (K) | $g$ | $\frac{dH_{c2}}{dT_c}$ (T/K) | $T_{c2B}$ (K) | $t_0 = \frac{\gamma_{c2B}^{2B}}{\gamma_{c2}^0}$ (nm) |
|-------------------|---------------------|-----------|-----|-----------------------------|---------------|-----------------------------------------------|
| $\lambda_{11}$    | $\lambda_{22}$      | $\lambda_{12}$ | $\lambda_{21}$ | 3.4 | 2.23 | -4.48 | 1.38 | 0.41 | 244 |
| $\lambda_{22}$    | 0.2                  | 0         | 0.2 | 0.2 | 2     | -0.532 | - | - | - |

| H-field direction | Band 1                  | Band 2                  |
|-------------------|-------------------------|-------------------------|
| $v_{ab}$ (m/s)    | $v_c$ (m/s) | $H_{c2B}^{2B}$ (T) | $H_{c2}^{c2}$ (T) | $\alpha_M$ | $v_{ab}$ (m/s) | $v_c$ (m/s) | $H_{c2B}^{2B}$ (T) | $H_{c2}^{c2}$ (T) | $\alpha_M$ |
| $ab$              | 18000                  | 115000                  | 1.29 | 6.31 | 2.23 | 0.82 | 1.38 | 0.41 | 244 |
| $c$               | 18000                  | 115000                  | 8.24 | 6.31 | 2.23 | 0.82 | 1.38 | 0.41 | 244 |

FIG. S4: One-band (1B) and two-band (2B) calculations for (a) $H \parallel ab$ and (b) $H \parallel c$.

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