

Upper limit on magnetic field expulsion in optically driven K_3C_{60} G. De Vecchi^{1,*}, M. Buzzi^{1,*}, G. Jotzu^{1,*}, S. Fava¹, T. Gebert¹, G. Magnani², D. Pontiroli², M. Riccò², and A. Cavalleri^{1,3,‡}¹Max Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany²Dipartimento di Scienze Matematiche, Fisiche e Informatiche, Università degli Studi di Parma, 43121 Parma, Italy³Department of Physics, Clarendon Laboratory, University of Oxford, Oxford OX1 3PU, United Kingdom

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Photoexcited K_3C_{60} displays several properties reminiscent of equilibrium superconductivity, including transient optical spectra, pressure dependence, and I - V characteristics. However, these observations do not decisively establish nonequilibrium superconductivity, which may be further evidenced by transient magnetic field expulsion measurements, as shown recently in driven $YBa_2Cu_3O_{6.48}$. Here, we search for transient magnetic field expulsion in K_3C_{60} by measuring Faraday rotation in a magneto-optic material placed in its vicinity. Unlike in the case of $YBa_2Cu_3O_{6.48}$, inhomogeneous, metallic K_3C_{60} powders reduce the size of the effect. With the ~ 50 nT magnetic field resolution achieved in our experiments, we provide an upper limit for the photoinduced diamagnetic volume susceptibility ($\chi_v > -0.1$). On this basis, we conclude that the photoinduced phase has weaker magnetic susceptibility than superconducting K_3C_{60} at zero temperature. Yet, from recent nonlinear transport measurements in this granular material, we expect a light-induced state similar to the equilibrium superconductor near $0.8 T_c$, for which $\chi_v > -0.1$. A definitive conclusion on the presence or absence of Meissner diamagnetism cannot be made for K_3C_{60} with the current resolution.

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I. INTRODUCTION

Optical manipulation of quantum materials has been extensively deployed in search of out-of-equilibrium functional phases, such as photoinduced ferroelectricity [1,2], magnetism [3–5], charge density wave order [6], topology [7,8], and superconductivity [9–15]. Among all these phenomena, the optical enhancement of superconductivity has attracted considerable attention.

In the specific case of K_3C_{60} , excitation with midinfrared and terahertz optical pulses results in a metastable nonequilibrium phase exhibiting optical conductivity spectra, characterized by a $1/\omega$ divergence of the imaginary conductivity (σ_2) and a gap in the real part (σ_1) [12] [Fig. 1(a)]. These features—reminiscent of those observed in the equilibrium superconducting state of K_3C_{60} —were observed up to base temperatures far exceeding the equilibrium superconducting critical temperature of the sample. Quantitative corrections to the retrieved optical constants, which also yield optical properties similar to those of superconducting K_3C_{60} measured in equilibrium for $T < T_c$, have nevertheless led to some

controversy [16,17]. This is ultimately rooted in the fact that the optical properties alone cannot definitively establish superconductivity, as these optical features are necessary but not sufficient conditions for superconductivity. A generic Drude conductor with very small scattering rate would have the same finite frequency conductivity as a superconductor (for frequencies larger than the scattering rate) and would show similar features in the optical conductivity spectra discussed above.

The pressure scaling of the transient optical conductivity [13], vanishing two-probe electrical resistance [14], and nonlinear current-voltage characteristics typical of a finite-temperature granular superconducting state [18,19] [Fig. 1(b)] further strengthen the case for superconductivity, although additional experimental probes are needed to clarify the underlying microscopic physics.

Here, we turn to a search for magnetic field expulsion in K_3C_{60} when the sample is photoexcited in a static magnetic field, as shown in recent experimental reports for a light-driven cuprate [20]. The immediate rationale for these experiments is that a nonsuperconducting photoinduced state in which the mobility is enhanced would not modify a static magnetic field. Conversely, if the material were turned promptly into a superconductor, the magnetic field would be dynamically expelled, in analogy with field-cooled Meissner diamagnetism at equilibrium [21].

II. OPTICAL FARADAY MAGNETOMETRY

To track the time- and space-dependent magnetic field surrounding a K_3C_{60} pellet after optical excitation, we

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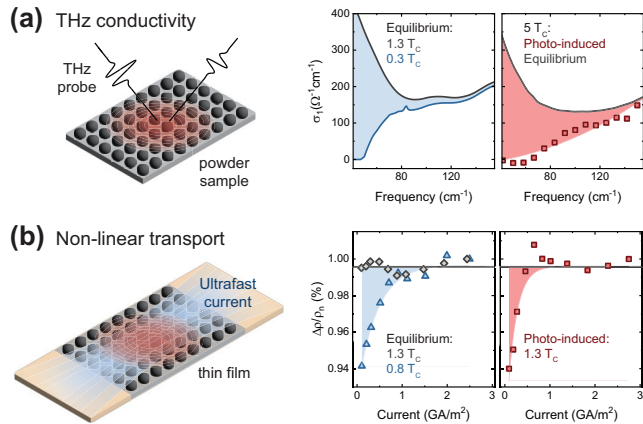


FIG. 1. Superconductinglike transport properties of photoexcited K_3C_{60} . (a) The terahertz conductivity spectra of a K_3C_{60} powder sample are measured using broadband THz pulses (left panel). The real part of the optical conductivity (right panels) is extracted both at equilibrium while cooling across the superconducting transition (dark gray for $T > T_c = 20$ K, blue for $T < T_c$) and out of equilibrium (dark gray line and red symbols refer to spectra measured before photoexcitation and after photoexcitation, respectively). A gap opens at equilibrium below T_c and after photoexcitation above T_c , indicating dissipationless transport. Data reproduced from Ref. [29]. (b) Ultrafast electrical transport is measured in a thin K_3C_{60} film using ultrafast current pulses. The right panel shows the nonlinear transport properties of the sample in the equilibrium superconducting state (blue triangles) when increasing the intensity of the ultrafast current pulse. Similar nonlinear properties are measured after midinfrared photoexcitation above T_c (red symbols). These nonlinear features are absent at equilibrium above T_c (gray diamonds). Data reproduced from Ref. [18].

employed a time-resolved magneto-optical imaging technique [20,22–24] [Fig. 2(a)].

This technique is analogous to the one used to measure a transient magnetic field expulsion in driven $YBa_2Cu_3O_{6.48}$ [20]. However, here the unperturbed metallic K_3C_{60} pellet adjacent to the photoexcited volume is expected to shield ultrafast changes in the local magnetic field, making the experiment more challenging. The presence of a time-dependent magnetic flux threading the metallic unperturbed pellet would induce an electromotive force at its boundary and generate eddy currents, counteracting the magnetic flux change through the material. This effect can be described by an equivalent LR low-pass circuit (where L is the inductance of the metallic K_3C_{60} pellet and R is its resistance). For the geometry relevant to this experiment, the L/R time constant is ~ 40 ps (see Appendix H). Hence, regardless of the detector's speed, the response time of magnetic field changes is intrinsically limited by the adjacent metallic sample. Note that this was not the case in the previous experiments [20] in a - c cut $YBa_2Cu_3O_{6.48}$, for which the low c -axis conductivity would strongly reduce a shielding current loop.

A ferrimagnetic $Lu_{3-x}Bi_xFe_{5-y}Ga_yO_{12}$ (hereafter Bi:LIGG) magneto-optic detector [25] was chosen for the present experiments due to its high magnetic field sensitivity. Given the inductive time constant discussed above, the ~ 100 ps [24,26–28] temporal response of this ferrimagnetic

detector is likely appropriate. Importantly, it is smaller than the lifetime of the metastable superconductinglike transport features observed in driven K_3C_{60} , lasting several nanoseconds [14].

We positioned the detector above the K_3C_{60} pellet, as shown in Fig. 2(a) and Appendixes A and B. Bi:LIGG encoded the local magnetic field z component into a polarization rotation of a linearly polarized probe pulse through the Faraday effect. The spot size of the probe laser limited the achieved spatial resolution. A pellet of ~ 1 mm diameter and ~ 30 μm thickness was prepared by pressing K_3C_{60} polycrystalline powders in an oxygen-free glovebox to avoid oxidation [Fig. 2(b) and Appendix C]. The pellet was sealed between the Bi:LIGG detector and a diamond window, transparent to the midinfrared excitation pulse (Appendix D). The entire device is schematically illustrated in [Fig. 2(a)]. A Helmholtz coil pair applied a 0.5 mT magnetic field along the z axis. A linearly polarized 800 nm ultrashort probe pulse, focused to ~ 50 μm FWHM, was directed at near-normal incidence onto the ~ 3 μm thick Bi:LIGG detector. After transmission through the Bi:LIGG, the detector's surface closest to the K_3C_{60} pellet reflected the pulse. Analyzing the probe's polarization yielded a measurement of the z -axis component of the local magnetic field, averaged over the volume traversed by the probe laser pulse in the magneto-optic detector. To enhance the intensity of the reflected probe beam and suppress noise from light scattering off the rough K_3C_{60} pellet surface, a ~ 150 nm thick layer of aluminum was deposited on the side of the magneto-optic detector closest to the K_3C_{60} pellet [Fig. 2(a) and Appendix D].

The externally applied magnetic field B_{app} was sinusoidally modulated in time, synchronously to the laser probe pulse train. Signals acquired with $-B_{app}$ were subtracted from those acquired with $+B_{app}$ (see Supplemental Material S2 [30]), distilling the magnetic field contributions to the polarization rotation and suppressing potential nonmagnetic contributions such as static birefringence.

III. EQUILIBRIUM MAGNETIC FIELD EXCLUSION

In these equilibrium magnetic field expulsion measurements, B_{app} was modulated in time, as the K_3C_{60} sample was held in its superconducting state. Therefore, the resulting magnetic response arose from the exclusion of a time-varying magnetic field rather than the expulsion of a static magnetic field. The generated magnetization is equivalent to that measured in zero-field cooling experiments. This effect is related to the perfect conductivity of a superconductor and is distinct from the Meissner effect (field-cooled magnetization). In general, magnetic field exclusion produces a larger signal than magnetic field expulsion as it reduces the impact of trapped flux [21] (see Supplemental Material S3 [30] and Appendixes E and F).

The colored shadings in Fig. 2(a) represent finite element simulations of the expected changes in the z component of the magnetic field surrounding the K_3C_{60} pellet. These simulations show a reduction in the magnetic field above the sample (blue regions) as the magnetic flux is excluded from it. The K_3C_{60} pellet was modeled as a homogeneous medium with a negative diamagnetic susceptibility $\chi \sim -0.5$, consistent with

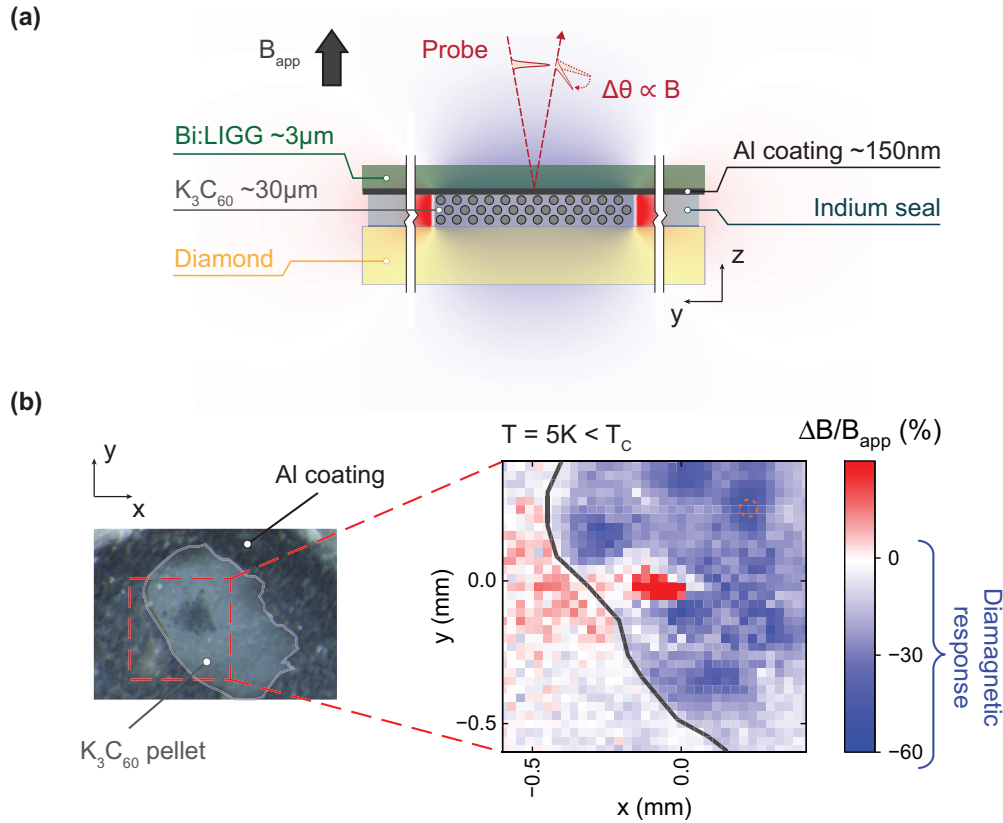


FIG. 2. Equilibrium Faraday magnetometry. (a) Sketch of the experimental geometry for Faraday magnetometry. To avoid oxidation, a pellet of pressed K_3C_{60} powder was sealed between a Bi:LIGG magneto-optic crystal and a diamond window. The local magnetic field surrounding the K_3C_{60} pellet was measured by tracking the Faraday polarization rotation of a linearly polarized 800 nm probe pulse, reflected after propagation through the Bi:LIGG crystal. A 0.5 mT magnetic field (B_{app}) was applied in the z direction, and its polarity was periodically cycled to isolate the magnetic contributions to the polarization rotation. A 150 nm thick aluminum coating was deposited on the Bi:LIGG surface to improve the signal-to-noise ratio (Appendix D). Additionally, the result of a finite element simulation (Appendix G) highlights the expected magnetic field changes induced by field expulsion in the superconductor (blue indicates reduction and red indicates enhancement of the local magnetic field). (b) (Left panel) Micrograph of the K_3C_{60} sample on the Al-coated magneto-optic detector. The gray line indicates the edges of the sample, and the dashed red line marks the region where the magnetic field was measured by raster scanning the sample relative to the probe beam. (Right panel) Two-dimensional map of the z component of the local magnetic field, measured as a function of the x and y positions of the probe at a constant temperature $T = 5\text{ K} < T_c$. An inhomogeneous reduction of the magnetic field is measured above the sample (blue), indicating the spatially inhomogeneous magnetic field shielding. The black line indicates the outline of the K_3C_{60} sample. The dashed circle indicates where the time-resolved experiments were carried out. The colormaps in panels (a) and (b) share the same color scale.

values typically measured in zero-field-cooled K_3C_{60} powders (Appendices F and G).

The panel on the left-hand side of Fig. 2(b) presents a micrograph of a K_3C_{60} pellet, with the red dashed box indicating the region investigated using optical magnetometry. This measurement was performed by raster scanning the device [Fig. 2(a)] relative to the laser probe beam. The panel on the right-hand side of Fig. 2(b) displays a two-dimensional map of the measured z component of the magnetic field when the K_3C_{60} pellet is cooled to $T = 5\text{ K}$, well into its superconducting state ($T_c \sim 20\text{ K}$). In this map, a gray line marks the sample edge for reference. A reduction in the local magnetic field (blue) is observed above the sample, in agreement with the simulations shown in Fig. 2(a).

Notably, the field map reveals significant spatial inhomogeneity in the degree of magnetic field exclusion across the pellet. Some regions show a field exclusion as high as $\sim 60\%$ (a value that becomes higher at lower values of the externally

applied field; Appendix F), while others show little to none. This result agrees with previous studies that reported granularity and spatial defects in K_3C_{60} [18,19,31].

IV. SEARCH FOR MAGNETIC FIELD EXPULSION IN OPTICALLY DRIVEN K_3C_{60}

The same K_3C_{60} pellet was then excited with a $\sim 1\text{ ps}$ long, $8\text{ }\mu\text{m}$ wavelength midinfrared pulse (Supplemental Material S1 [30]), which is known to induce metastable superconductinglike THz transport properties [14] and other electrical features discussed above. Figure 3(a) illustrates the experimental geometry, where the photoexcited region (marked in red) has lateral dimension and thickness equal to the midinfrared pump beam cross section and its penetration depth in K_3C_{60} , respectively (Appendix H).

If a diamagnetic response emerges following photoexcitation in K_3C_{60} , the resulting spatial profile changes in the

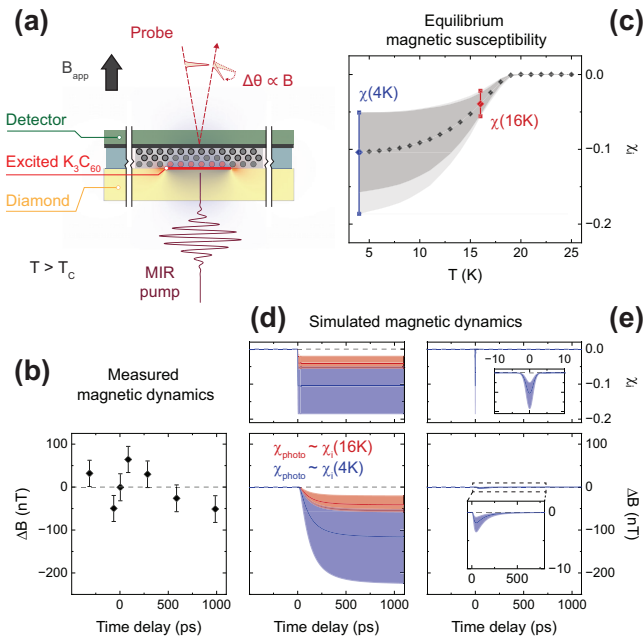


FIG. 3. Search for magnetic expulsion in photoexcited K_3C_{60} . (a) Sketch of the experimental geometry [same as in Fig. 2(a)]. The sample is at a base temperature $T = 50 \text{ K} > T_c$ and excited with a midinfrared pump pulse ($\lambda \sim 8 \mu\text{m}$). A possible pump-induced diamagnetic response would manifest as a magnetic field reduction at the detector position (blue shading of the overlaid simulation). The pump-induced changes in the local magnetic field were quantified using Faraday magnetometry in a Bi:LIGG detector as a function of pump-probe time delay. A 0.5 mT magnetic field (B_{app}) is applied in the z direction. Its polarity is periodically cycled to isolate the magnetic contributions to the rotation of the polarization. (b) Time dependence of the pump-induced changes in the z component of the local magnetic field ΔB . The error bars denote the standard error of the mean. (c) Equilibrium temperature dependence of the diamagnetic susceptibility χ_i at 1 mT applied field of six pellets of K_3C_{60} , similar to the one shown in Fig. 2(b). The dark gray diamonds represent the average diamagnetic susceptibility, the gray band represents the interval within one standard deviation from the average, and the light gray band represents the range between the measured maximum and minimum values (see Appendix F). (d) Steplike quench of the diamagnetic susceptibility (top panel) in the photoexcited region at time zero such that $\chi_{\text{photo}}(t \rightarrow +\infty) = \chi_i(4 \text{ K})$ (blue) and $\chi_{\text{photo}}(t \rightarrow +\infty) = \chi_i(16 \text{ K})$ (red). The bottom panels indicate the respective simulated magnetic field dynamics. Shaded bands indicate the range between the maximum and minimum values of χ_i measured at that temperature. (e) Same as in panel (d) but for $\chi_{\text{photo}}(t)$ following a Gaussian pulse peaking at $\chi_{\text{photo}}(t = 0) = \chi_i(4 \text{ K})$.

magnetic field surrounding the photoexcited region should resemble the finite element simulations displayed in Fig. 3(a). Specifically, a reduction of the local magnetic field should be observed above the center of the photoexcited region.

The pump and the probe beam were positioned at a location in the K_3C_{60} pellet for which the best field exclusion was observed in the equilibrium measurements [dashed circle in Fig. 2(b)]. The sample was then photoexcited at a fluence of 40 mJ cm^{-2} , starting from its normal metallic state at $T = 50 \text{ K} > T_c = 20 \text{ K}$.

Figure 3(b) displays the measured magnetic dynamics as a function of pump-probe delay for an externally applied field $B_{\text{app}} = 0.5 \text{ mT}$. Within the current sensitivity of our experimental setup, no statistically significant signal larger than $\sim 50 \text{ nT}$ could be detected up to 1 ns time delay.

This result should be evaluated by comparing it with the magnetic field dynamics generated by a disk of the same size as the photoexcited volume acquiring a finite intrinsic diamagnetic susceptibility χ_i after photoexcitation. As a possible value for χ_i , we take that measured in K_3C_{60} pellets at equilibrium after field cooling. Without knowledge about the granularity and the density of defects in the photoexcited state and given the significant sample-to-sample variability, we used statistical values extracted from a set of pellets similar to the one considered here to estimate χ_i . We measured the field-cooled intrinsic equilibrium diamagnetic susceptibility of six pellet samples using a commercial magnetometer (Appendix F). The temperature dependence of χ_i averaged across all samples is displayed in Fig. 3(c) (filled diamonds). The shaded bands represent the standard deviation of the sample population (gray) and the range between maximum and minimum values at each temperature (light gray).

Figures 3(d) and 3(e) show simulations of the magnetic field dynamics generated by a medium with a pump-induced, time-dependent magnetic susceptibility, obtained by solving time-dependent Maxwell's equations. The 150 nm thick aluminum layer was modeled as a metal with $\sigma_0 = 3 \times 10^5 \Omega^{-1}\text{cm}^{-1}$. A 1 mm diameter, 35 μm thick disk with a dc conductivity $\sigma_0 = 800 \Omega^{-1}\text{cm}^{-1}$ was used to reproduce the unperturbed, metallic K_3C_{60} pellet measured. The photoexcited volume was modeled as a 100 μm diameter disk (matching the size of the pumped spot), with a thickness of 400 nm (equal to the electric field penetration depth at the pump center frequency). Its magnetic response was modeled assuming a homogeneous, time-dependent magnetic susceptibility $\chi_{\text{photo}}(t)$ (Appendix H).

We considered two different scenarios for $\chi_{\text{photo}}(t)$: (1) a metastable diamagnetic response emerging after photoexcitation, as measured by the transient conductivity experiments and (2) a diamagnetic response persisting only for the duration of the drive, to test the level of uncertainty on the resolution of our measurement.

In the first scenario, $\chi_{\text{photo}}(t)$ is modeled as a negative step function with $\sim 1 \text{ ps}$ fall time reaching values equal to the average field-cooled magnetic susceptibility of the K_3C_{60} pellets at 4 and 16 K (blue and red lines in Fig. 3(d), top panel, respectively). The shaded areas represent the range between the maximum and minimum values within the sample population at these temperatures.

The lower panel of Fig. 3(d) shows the corresponding magnetic field changes. Regardless of the final value $\chi_{\text{photo}}(t \rightarrow +\infty)$, the traces show a long-lived magnetic field change after an initial reduction with a time constant of $\sim 150 \text{ ps}$. This slower drop is due to the inductive response of the unperturbed metallic K_3C_{60} pellet and the thin aluminum film deposited on the Bi:LIGG detector (Appendix H). Given this timescale imposed by inductive currents, the response of the magnetization in the Bi:LIGG magneto-optic detector ($\sim 150 \text{ ps}$ [24]) does not play a role in determining the measured signal dynamics. The long-lived magnetic field change

is estimated to be ~ -115 nT for $\chi_{\text{photo}}(t \rightarrow +\infty) = \chi_i(4 \text{ K})$ and ~ -40 nT for $\chi_{\text{photo}}(t \rightarrow +\infty) = \chi_i(16 \text{ K})$.

In the second scenario, $\chi_{\text{photo}}(t)$ follows a Gaussian pulse peaking at $\chi_{\text{photo}}(t = 0) = \chi_i(4 \text{ K})$ [Fig. 3(e), top panel]. Due to the inductive response of nearby metallic elements, the maximum amplitude of the magnetic field change is severely reduced to ~ -2 nT [Fig. 3(e), lower panel], well below our experimental resolution.

Given the results of these simulations and the ultimate sensitivity of our measurements [~ 30 nT, as determined by the standard error of the mean shown in Fig. 3(b)], we would be able to detect a light-induced magnetic field change only if photoexcited K_3C_{60} entered a metastable state with a diamagnetic susceptibility equal to that measured in the 4 K equilibrium superconducting state. We note that the nonlinear current measurements of Fig. 1(b), performed under similar excitation conditions, showed that the measured superconducting-like electrical features are equivalent to those seen at equilibrium close to T_c ($T = 16 \text{ K}$) [18]. According to our time-dependent simulations, the present experimental resolution is not sufficient to detect a diamagnetic susceptibility corresponding to the equilibrium 16 K response.

V. DISCUSSION

The present experiment sets a limit to the maximum magnetic field expulsion that could be observed in driven K_3C_{60} using the best ultrafast magnetometry technique achievable to date. The experimental conditions are unfavorable due to the geometry of the photoexcited state, its distance from the magneto-optic detector, and the inductive response of the unperturbed metallic K_3C_{60} . Although we achieved sensitivity to magnetic field changes of a few tens of nanotesla, the upper limit we can set for the magnitude of the diamagnetic susceptibility of the excited region is of order -10^{-1} . This value is relatively high, even for an equilibrium type-II superconductor, but lower than the value measured in driven $\text{YBa}_2\text{Cu}_3\text{O}_{6.48}$ [20]. For reference, the magnitude of the equilibrium magnetic susceptibility of some K_3C_{60} samples we measured is lower than this limit, even at temperatures as low as $0.2 T_c$ [Fig. 3(c)].

Furthermore, it is becoming clear that quasiparticles are present in the photoexcited state [16–19,32] and that granularity and defects are an important feature in K_3C_{60} . Therefore, we would expect a photoinduced diamagnetic response to be weaker than that of its low-temperature equilibrium counterpart.

The development of more sensitive, time-resolved magnetometry techniques or advances in sample preparation could further improve the sensitivity of this experiment. Additionally, performing this measurement at 10 THz excitation frequency might quantitatively improve the characteristics of the photoexcited state [29].

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DATA AVAILABILITY

The data that support the findings of this article are openly available in the following repository [33].

APPENDIX A: ULTRAFAST MAGNETOMETRY TECHNIQUE

Ultrafast optical magnetometry relies on the Faraday effect, which directly relates the magnetic field applied to a material to the polarization rotation of a linearly polarized beam traversing the medium. This relation is usually reported as

$$\theta = V \cdot \int_0^L B(z) dz, \quad (\text{A1})$$

where θ represents the rotation of the polarization of the input beam, $B(z)$ is the magnitude of the magnetic field along the light propagation direction inside the medium, and L is the thickness of the medium. The proportionality constant V is known as the Verdet constant, a material-dependent constant that also depends on other parameters, such as the wavelength of the incoming polarized light. This expression highlights the fact that the magnetic field is averaged across the thickness of the material traversed. Therefore, if B varies strongly over distances of order $\sim L$, its spatial dependence is blurred in the accumulated polarization rotation.

In the past, the Faraday effect in ferromagnetic crystals and thin films has been used to image the magnetic properties of superconductors at equilibrium [28,34]. These types of detectors (such as $\text{Bi}:\text{R}_3\text{Fe}_5\text{O}_{12}$, EuS , and EuSe) offer very high sensitivity ($V \sim 10^5 \text{ rad T}^{-1} \text{ m}^{-1}$) but have limited time resolution, down to 100 ps at best [24], due to the presence of low-lying magnetic excitations (e.g., ferromagnetic resonance) at sub-THz frequencies. On the other hand, diamagnetic II–VI and III–V semiconductors such as ZnSe , ZnTe , and GaP have a magneto-optic response featuring Verdet constants that are two to three orders of magnitude smaller than those observed in ferromagnetic materials but have the advantage of not being magnetically ordered and offer significantly better time resolution [20,22,24,35].

Given the high conductance of the K_3C_{60} pellet measured in this work, picosecond time resolution was unnecessary (see Appendix H), and we opted for an improved magnetic field sensitivity. The measurements shown throughout the manuscript were performed using a $\text{Lu}_2\text{Bi}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$ ($\text{Bi}:\text{LIGG}$, see Appendix B) magneto-optic crystal. Its sensitivity to magnetic fields was calibrated before each measurement in the exact experimental conditions used for the measurement. We measured the amount of angular polarization rotation in a position of $\text{Bi}:\text{LIGG}$ far from the sample (see Fig. 2), where the magnetic field was equal to the uniform externally applied magnetic field. In this way, we could extract

a calibration factor specific to our experimental conditions, which allowed us to convert the measured polarization rotation into the value of the local magnetic field in the crystal.

Finally, to have an accurate calibration of the actual value of the magnetic field, the current-to-magnetic field constant of the Helmholtz coil pair was independently calibrated *in situ* using a Lakeshore 425 gaussmeter. These calibration measurements yielded a Verdet constant of $\sim 0.5 \times 10^5 \text{ rad T}^{-1} \text{ m}^{-1}$ for Bi:LIGG (Appendix B).

APPENDIX B: DETECTOR CHARACTERIZATION

The $\sim 3 \mu\text{m}$ thick Bi-substituted lutetium iron gallium garnet ($\text{Lu}_2\text{Bi}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$, Bi:LIGG for short in the following) magneto-optic detector was obtained through a commercial supplier and grown by Liquid-Phase Epitaxy on a (100)-oriented, $500 \mu\text{m}$ thick $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ crystal (GGG). The composition of the Bi:LIGG sample was measured using energy-dispersive x-ray spectroscopy, yielding the following percentage composition of elements: O 58.98%, Fe 19.43%, Ga 5.99%, Lu 10.28%, and Bi 5.32%. This result is in good agreement with the chemical formula reported above.

Measurements reported in another manuscript published by some of the authors [24] reveal the presence of a low-frequency magnon (centered at a frequency of $\sim 6 \text{ GHz}$) in the same Bi:LIGG compound used here as a magneto-optic detector. The time response of the material's magnetization, and consequently the response of the Faraday polarization rotation, is limited to the inverse of this excitation frequency $\sim 160 \text{ ps}$. This time resolution does not constitute an issue for the measurement we want to perform here due to the intrinsic low-pass filtering effect due to the magnetic shielding from the metallic K_3C_{60} pressed pellet (Appendix H).

APPENDIX C: SAMPLE GROWTH, CHARACTERIZATION, AND PREPARATION

The K_3C_{60} powder used for the pellets discussed in this work was prepared and characterized as reported previously [12–14,29]. Stoichiometric amounts of ground C_{60} powder and potassium were placed in a sealed Pyrex vial, which was evacuated to 10^{-6} mbar pressure. While keeping the C_{60} powder and solid potassium separated, the vial was kept at 523 K for 72 h and then at 623 K for 28 h so that the C_{60} powder was exposed to pure potassium vapor. The vial was then opened inside an Ar glovebox ($<0.1 \text{ ppm O}_2$ and H_2O), where the powder was reground and pelletized before annealing at 623 K for 5 days. X-ray diffraction measurements were then carried out on the resulting K_3C_{60} powder, confirming that it was phase pure, with an average grain size between 100 and 400 nm (Fig. 4). The quality of the sample was then checked by producing a pellet (see discussion below) and measuring its magnetic susceptibility (see Appendix F). These measurements revealed a significant variability across the different pellets measured, even if the pellet preparation process was reproduced consistently for each measurement.

This K_3C_{60} powder was then used to prepare the pellet measured in the main text. The powder was pressed between

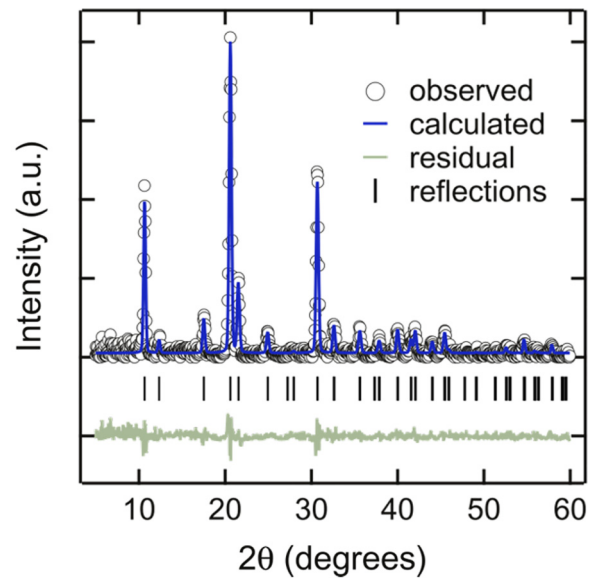


FIG. 4. X-ray diffraction data and single fcc phase Rietveld refinement for the K_3C_{60} powder used in this work.

two press dies in a polypropylene mask to control the resulting pellet's thickness and shape (Fig. 5).

Additionally, the surface of each press die was covered with Kapton foil, which prevented the powder sample from sticking to the metallic surface after pressing. The whole sample preparation procedure was performed inside an Ar glovebox ($<0.1 \text{ ppm O}_2$ and H_2O) to avoid oxidization of the sample. All the tools and surfaces coming in contact with the sample were cleaned with acetone in an ultrasonic bath, rinsed with isopropanol, and baked for some minutes on a hot plate before being carried inside the glovebox to avoid alteration of the K_3C_{60} properties. An example of a pellet resulting from this process is shown in Fig. 5(b). The sample shown in Fig. 2 has been prepared with the same procedure. Its irregular shape is due to the edges breaking when removing the pellet from the polypropylene mask.

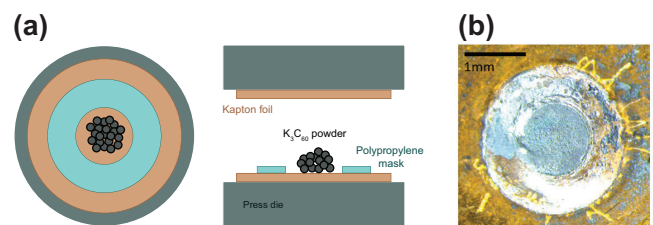


FIG. 5. Process to produce a K_3C_{60} pellet of known thickness. (a) Sketch representing the preparation phase. Some K_3C_{60} powder is placed in the center of a polypropylene ring $30 \mu\text{m}$ thick on top of a Kapton foil disk. This disk prevents the K_3C_{60} powder from sticking to the metallic surface of the press die. The top view is on the left panel, while the side view is on the right panel. (b) Micrograph showing a 1 mm diameter pellet after pressing the powder in the polypropylene mask.

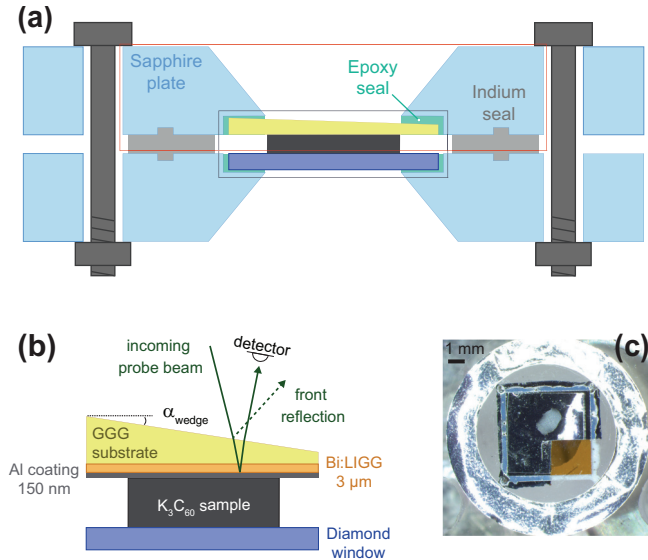


FIG. 6. (a) Schematic representation of a section of the sample holder. The substrate of the magneto-optic detector (yellow) and the diamond window (dark blue) were each glued to a sapphire plate using sapphire-based epoxy glue. The K₃C₆₀ sample was placed between the magneto-optic detector and diamond window, touching them to ensure thermal contact. A ring of indium [see panel (c)] was pressed between the two sapphire plates to seal the sample. The preparation of this sample holder was performed inside an Ar glove-box to avoid sample contamination. (b) Side view of the experimental geometry. The GGG substrate of the Bi:LIGG detector was wedged at an angle $\alpha_{\text{wedge}} \sim 1.5^\circ$ (Appendix B) to separate the front and back reflections from each other spatially and only collect the latter with our photodetector. (c) The micrograph shows the front view of the upper sapphire plate with the magneto-optic detector glued to it and coated with aluminum. The indium ring with an internal diameter of 8 mm used to seal the sample is also visible. The sample in the center is the same as in Fig. 2.

APPENDIX D: SAMPLE HOLDER AND EXPERIMENTAL GEOMETRY

Figure 6 shows the sample holder panel (a) and the geometry of the experiment panel (b). A good thermal contact of the sample was ensured by pressing the K₃C₆₀ pellet between the magneto-optic detector and the diamond window. Both the window and the detector were then glued to a sapphire plate using sapphire-based epoxy glue, which had good thermal conductivity and poor electrical transport properties. Finally, the sapphire plates were thermally contacted with a cold finger to cool down the sample holder. The Bi:LIGG detector was coated with a 150 nm thick aluminum layer to maximize the intensity of the light propagating through the magneto-optic crystal and to suppress diffuse scattering from the rough K₃C₆₀ surface that would have caused additional noise. Part of the detector was not coated with aluminum [Fig. 6(c)]. We used the sharp edges of the Al coating to perform knife-edge scans and precisely identify the position and size of the probe beam *in situ*. Furthermore, by letting the intense midinfrared pump propagate with the probe beam in the uncoated region, we could find the time overlap between the pump and the probe. Conversely, the sample and the metallic film com-

pletely prevented this cross-talk between the pump and probe by absorbing or reflecting the pump radiation. The sample holder was designed to minimize the presence of electrically conductive loops, in which eddy currents would build up to shield rapid changes of the magnetic field (Appendix E). The only highly conductive components close to the photoexcited region are the puck of pressed K₃C₆₀ powder, which is metallic with a conductivity of $\sim 10^5 \text{ S m}^{-1}$, the aluminum coating deposited on the Bi:LIGG detector, and the sealing indium ring shown in Fig. 6(c). We discuss the effect of the metallic K₃C₆₀ puck and the aluminum film in Appendix H. The presence of the indium ring can be neglected. Due to its poor mutual inductance with the much smaller photo-excited region, this indium ring shields time-dependent changes of magnetic field generated by the photoexcited region by less than one part per million.

APPENDIX E: SHIELDING OF TIME-VARYING MAGNETIC FIELDS

This Appendix elaborates on how systems with high electrical conductivity shield time-varying magnetic fields, as the Faraday-Lenz law describes, in the experimental configurations relevant to this manuscript. This concept applies to superconductors below T_c and high-conductivity metals.

1. In superconductors

Magnetic field exclusion occurs when a superconductor is cooled below T_c , and a time-varying magnetic field is applied. This phenomenon is due to the perfect conductivity of the YBa₂Cu₃O₇ sample below T_c . Magnetically induced persistent shielding currents keep the magnetic flux constant in the superconductor, as the Faraday-Lenz law prescribes. The magnetic field is entirely excluded from the sample volume, even if defects exist, unlike what happens for the Meissner effect (Supplemental Material S3 [30]). This shielding response is observed in zero-field-cooled experiments [21,36].

Figure 7 qualitatively explains the equilibrium field dynamics in our experiment, which is very similar to the zero-field-cooled regime. Figure 7(a) shows the temperature evolution during a hypothetical experiment in which a sample is progressively cooled below T_c . Figure 7(b) shows the probe pulses used to sample the magnetic field at a specific point in time. When there is no sample, we expect to measure a value equal to the amplitude of the applied sinusoidal magnetic field. As described in Supplemental Material S2 [30], the magnetic field is extracted as the difference between the polarization rotation measured when the polarity of the sinusoidal wave is positive minus the one measured when the polarity is negative. Figure 7(d) schematically shows the evolution of the magnetic field upon cooling our sample across T_c . Initially, when the sample is above T_c , the magnetic field measured is almost equal to the applied magnetic field due to the higher resistivity of the sample in the normal state. As described above, the superconductor begins to partially shield the time-varying applied field after cooling across T_c . In this temperature regime close to T_c , the shielding is incomplete due to residual dissipation and the superconductor's low critical current density. Upon further cooling, the

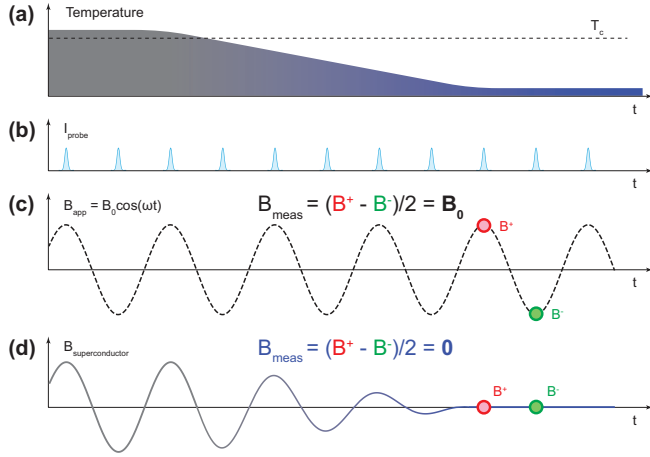


FIG. 7. (a) Sketch of the temporal profile of the temperature when cooling the sample below T_c . (b) Arrival time of the probe pulses sampling the magnetic field. (c) Externally applied magnetic field with a sinusoidal time evolution. (d) Magnetic shielding of the superconductor when cooling across T_c .

shielding improves as the superfluid density increases and the superconductor behaves like a perfect metal in which the eddy currents induced by the applied field become persistent due to the absence of dissipation. In this regime of perfect screening, the field on top of the sample is constant. Our differential measurement would yield a value of the magnetic field close to zero above the center of the superconductor. In practice, the magnetic field shielding of our pellet samples is incomplete (see the zero-field-cooled magnetization reported in Appendix F). Therefore, we measure a sizable but partial reduction of the externally applied magnetic field on top of the superconducting patches of our sample [Fig. 2(b)].

2. In metals

Metals' response to time-varying magnetic fields is similar to that of superconductors. The main difference is that their conductivity is finite, and electrical transport is dissipative. Therefore, the shielding currents induced in metals by time-varying magnetic fields decay with a characteristic time constant inversely proportional to their resistivity and are not persistent like in superconductors. Due to their geometry and relatively low conductivity, the decay times of eddy currents in the metallic elements in our sample holder (Appendix D) are much faster than the timescales of a few hundred Hz at which we switch the externally applied magnetic field. Therefore, the applied field changes are adiabatically slow for these currents and cause a negligible shielding response. Conversely, if we focus on the opposite limit, in which magnetic field changes happen on timescales much shorter than the decay time of eddy currents, the metal behaves similarly to a superconductor, as described in the previous subsection. That is why a Meissner response of the photoexcited state happening on ultrafast timescales would be almost entirely shielded by the metallic K_3C_{60} puck not excited by the pump [Appendix H and Figs. 3(d) and 3(e)].

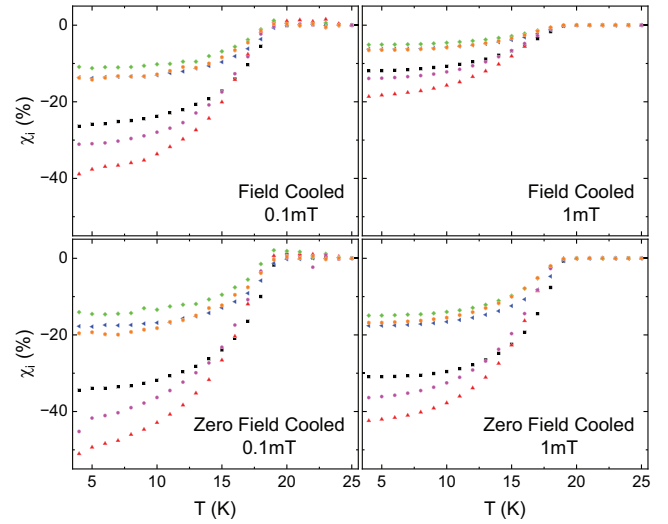


FIG. 8. Intrinsic (corrected for the demagnetizing factor of the sample) magnetic susceptibility χ_i extracted from SQUID magnetization measurements performed on six different samples both in field-cooled and zero-field-cooled conditions at 0.1 and 1 mT externally applied magnetic field.

APPENDIX F: EQUILIBRIUM MAGNETIC PROPERTIES OF K_3C_{60} PELLETS

This Appendix discussed the equilibrium magnetic properties of K_3C_{60} pellets. The data shown here have been measured with the experimental setup described in this manuscript and with a Quantum Design MPMS3 Superconducting Quantum Interference Device (SQUID). The two main characteristics that emerge from measurements performed on multiple samples are their poor quality compared to single crystals of K_3C_{60} and a broad sample-to-sample variability.

1. Sample variability

We performed a series of SQUID magnetometry measurements at equilibrium to test the quality of the K_3C_{60} powder, the possible effects of the pellet preparation procedure on the sample, and the overall reproducibility of our samples. We produced each pellet in an Ar glovebox, achieving disk-shaped samples similar to the one shown in Fig. 5(c), which had a known diameter and thickness. While still in the glove box, we encapsulated each pellet between two 50 μm thick indium sheets pressed together to protect it from contamination during the transfer to the SQUID. The magnetic field of the SQUID was applied parallel to the disk's axis. We accounted for demagnetizing effects using the approximated expression for the demagnetizing factor of a thin cylinder with the magnetization aligned along its axis: $N \sim 1 / (1 + 1.6/A)$, from reference [37]. In this formula, A is the ratio between the diameter and the height of the cylinder.

In Fig. 8, we report the intrinsic volume magnetic susceptibility extracted from data collected from six pellets produced with different batches of K_3C_{60} powder. We see a marked sample-to-sample variability and notice that none of the samples measured showed a complete magnetic field expulsion

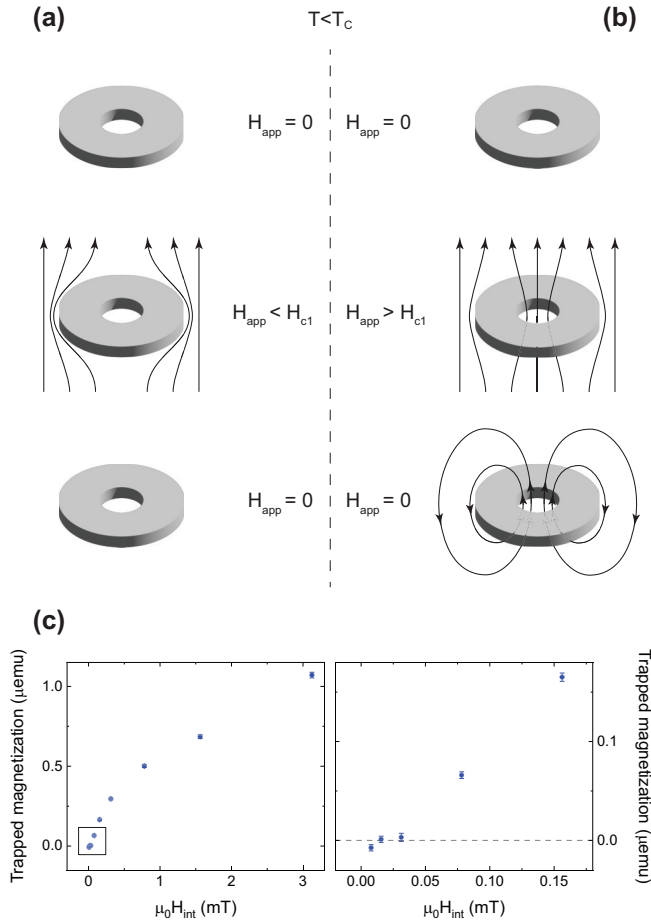


FIG. 9. (a), (b) Experimental protocol for the “trapped magnetic-moment” measurements described in Ref. [31]. When the externally applied magnetic field exceeds the first critical field (H_{c1}), the sample retains a trapped magnetic moment upon nulling again the applied field (b). (c) Trapped magnetic moment measured with a SQUID magnetometer as a function of the applied magnetic field corrected for demagnetizing effect (H_{int}) at 5 K. The right panel shows a zoom-in at low magnetic fields.

(“Field Cooled”) or exclusion (“Zero Field Cooled”) even at the lowest temperature and magnetic field applied. For this reason, we further investigated the equilibrium properties of our K_3C_{60} pellets, as described in the following subsection. The statistical quantities reported in Fig. 3(c) have been extracted from the “Field Cooled 1 mT” dataset shown in Fig. 8, as a proxy for the hypothetical magnetic field expulsion of the photoexcited state in the 0.5 mT externally applied field in our experiment.

2. Sample granularity

We performed magnetization measurements versus the applied magnetic field to investigate further our samples’ incomplete magnetic field expulsion (Fig. 8). We used a measurement protocol already employed to quantify the first critical field of K_3C_{60} single crystals in Ref. [31]. The idea of this method is displayed in Figs. 9(a) and 9(b).

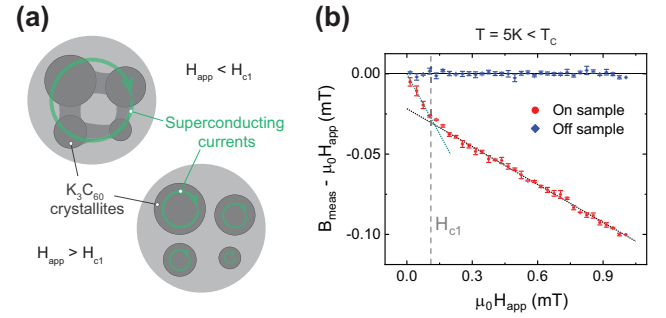


FIG. 10. (a) Schematic picture of the sample granularity and how it affects the sample’s magnetic properties. Panel (b) shows equilibrium magnetization measurements of the K_3C_{60} pellet displayed in Fig. 2, performed with the setup described there. The sample’s magnetization ($B_{meas} - \mu_0 H_{app}$) changes its slope at an amplitude equal to 120–130 μT of externally applied magnetic field, oscillating at 450 Hz. This change of slope indicates the crossing of the first critical field in the sample.

The sample is cooled from above T_c in zero magnetic field to a temperature lower than T_c . An externally applied magnetic field is then ramped up to a specific value, x , and set to zero again. At this point, the magnetic moment trapped inside the sample is measured. Suppose the value of the externally applied magnetic field is lower than the first critical field of the sample. In that case, no magnetic flux will penetrate the sample, and no trapped magnetic moment will be measured [Fig. 9(a)].

Conversely, if the applied field exceeds the first critical field, some magnetic flux will penetrate the sample’s volume. Upon nulling the externally applied field again, some superconducting currents will build up to keep the magnetic flux threading the sample volume constant (due to the Faraday-Lenz law), producing a trapped magnetic moment in the sample [Fig. 9(b)]. The sample is then heated above T_c to erase its memory, and the cycle is repeated by applying a different maximum value of the external magnetic field.

Figure 9(c) shows the trapped magnetic moment of a K_3C_{60} pellet at a temperature of 5 K for different values of the applied magnetic field corrected for the demagnetizing effects to achieve the internal field H_{int} . In this sample, the first critical field $\mu_0 H_{c1}$, defined above, lies between 30 and 80 μT .

Figure 10(b) shows the equilibrium field-dependent magnetization of the K_3C_{60} pellet in Fig. 2. These data have been acquired in a separate measurement using the same optical magnetometry setup [Fig. 2(a)]. Upon increasing the externally applied magnetic field past a critical value, the slope of the magnetization changes, indicating a change in the magnetic susceptibility of the sample. This critical value lies around 130 μT , comparable to the results of the “trapped magnetic-moment” measurements discussed above, and we identify it with H_{c1} of this sample. For comparison, the lower critical field extracted by Buntar *et al.* [31] from similar trapped magnetic-moment measurements was 1.2 mT in monocrystalline samples, which showed a magnetic susceptibility as good as -100% .

Figure 11 shows two-dimensional maps of the z component of the local magnetic field measured at $T < T_c$ above the

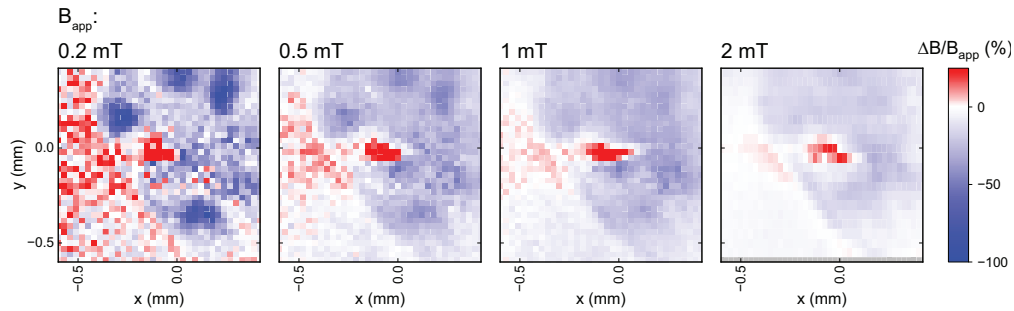


FIG. 11. Two-dimensional maps of the z component of the local magnetic field, measured as a function of the x and y positions of the probe at a constant temperature $T = 5 \text{ K} < T_c$ above the sample shown in Fig. 2(b) and with the same experimental setup [shown in Fig. 2(a)]. These data have been acquired for different values of the externally applied magnetic field. An inhomogeneous reduction of the magnetic field is measured above the sample (blue), indicating the spatially inhomogeneous magnetic field shielding. The shielding fraction ($\Delta B/B_{\text{app}}$) and the shielding inhomogeneity of the sample increase as the externally applied magnetic field is reduced. At the lowest applied field ($B_{\text{app}} = 0.2 \text{ mT}$), the minimum shielding fraction measured is as low as -85% . The red spot within the sample contour is due to an imperfection of the magneto-optic detector. The data acquired at $B_{\text{app}} = 0.5 \text{ mT}$ are the same as shown in Fig. 2(b).

sample shown in Fig. 2(b) and with the magnetometry setup of Fig. 2(a). The degree of spatial inhomogeneity and the magnitude of the sample's magnetic field shielding (blue) vary with the value of the externally applied magnetic field, which oscillates at 450 Hz (Appendix E). At the lowest applied field ($B_{\text{app}} = 0.2 \text{ mT}$), the measured shielding fraction ($\Delta B/B_{\text{app}}$) reached values as low as -85% , while at the highest field ($B_{\text{app}} = 2 \text{ mT}$), it is at best -30% . For low values of the applied magnetic field, patches appear in the sample where the magnetic shielding is significantly higher than the one measured at high values of the applied field.

These observations suggest that our powder samples are granular, with small superconducting grains connected by weak links, which break down at very low applied magnetic fields [Fig. 10(a)]. Moreover, these superconducting grains seem to be connected by weak links whose strength varies spatially throughout the sample, as suggested by the formation of macroscopic islands with a lateral size of $\sim 100 \mu\text{m}$ at low values of the externally applied magnetic field (Fig. 11). These hypotheses would explain the reduced diamagnetic susceptibility of the pellet samples and their much lower critical field compared to the literature values for single crystals. Similar conclusions agree with the results of transport measurements performed on thin films of K_3C_{60} [18,19].

The strong dependence of the equilibrium properties of K_3C_{60} on the externally applied magnetic field leads us to imagine that the photoexcited state also possesses a nontrivial susceptibility to the applied field. We address this question in Supplemental Material S4 [30], showing the results of pump-probe measurements obtained at various values of the externally applied magnetic field.

APPENDIX G: SIMULATED SPATIAL MAGNETIC FIELD DISTRIBUTION

The simulated distribution of the z component of the magnetic field around the whole K_3C_{60} pellet at equilibrium and around the photoexcited region is shown in Figs. 2(a) and 3(a). They have been calculated using a magnetostatic model in *comsol*. We modeled the superconductors as disks with a uniform diamagnetic susceptibility. The magnetic field generated

by the samples was calculated using *comsol* multiphysics to solve Maxwell's equations.

An aspect ratio (diameter/thickness) of 6 has been used to simulate the equilibrium sample, which does not respect the aspect ratio of the entire puck ($\sim 1 \text{ mm}/30 \mu\text{m} \sim 33$). However, this value is closer to the superconducting patches measured in reality [Fig. 2(b)], which had a lateral size of $\sim 200 \mu\text{m}$. Conversely, the pumped region was simulated as a thin disk with a diameter of $100 \mu\text{m}$ and a thickness of 400 nm , corresponding to the pump spot size and the pump penetration depth, respectively (Appendix H). The extreme aspect ratio of this geometry limits the changes to the applied magnetic field of the sample.

These simulations indicate that a sample developing a (transient) diamagnetic susceptibility would reduce the magnetic field measured with our probe on top of it.

APPENDIX H: SIMULATED MAGNETIC DYNAMICS

This Appendix discusses the time-dependent finite element simulations we performed to calculate the dynamics of the possible magnetic field expulsion from the photoexcited sample in our experimental configuration (Appendix D). We used *comsol* multiphysics to solve Maxwell's equations and evaluate the average magnetic field in a domain corresponding to the volume of the magneto-optic detector traversed by our probe pulse. This domain was $3 \mu\text{m}$ thick (as the Bi:LIGG crystal), had a diameter of $50 \mu\text{m}$ (equal to the FWHM of the probe beam), and was placed a distance of $35 \mu\text{m}$ from the excited region to account for the thickness of the K_3C_{60} puck. The pumped region was simulated as a thin disk with a diameter of $100 \mu\text{m}$ (approximately equal to the FWHM of the midinfrared pulse) and a thickness of 400 nm [29].

To faithfully simulate the dynamics of the local magnetic field around the photoexcited region, we also considered the effect of the 150 nm thick aluminum layer coating the Bi:LIGG crystal and the $30 \mu\text{m}$ thick, 1 mm diameter unperturbed K_3C_{60} pellet. Due to their high conductivity, they act as a low-pass filter, reducing a possible signal generated by the sample (Appendix E). To keep the size of the simulation domain manageable, we limited the diameter of the

TABLE I. Calculated long-lived magnetic field change ΔB following a metastable quench of the photoinduced diamagnetic susceptibility $\chi_{\text{photo}}(t)$ to values equal to the maximum, average, and minimum measured equilibrium magnetic susceptibility χ_i at 4 and 16 K (Appendix F).

$T = 4 \text{ K}$		$T = 16 \text{ K}$	
$\chi_{\text{photo}}(t \rightarrow +\infty)$	$\Delta B \text{ (nT)}$	$\chi_{\text{photo}}(t \rightarrow +\infty)$	$\Delta B \text{ (nT)}$
$\chi_{i, \text{max}} = -0.05$	-50	$\chi_{i, \text{max}} = -0.02$	-20
$\chi_{i, \text{ave}} = -0.105$	-115	$\chi_{i, \text{ave}} = -0.04$	-40
$\chi_{i, \text{min}} = -0.185$	-230	$\chi_{i, \text{min}} = -0.055$	-60

thin aluminum film to 2 nm, slightly smaller than the actual dimension in the experiment [Fig. 6(c)]. Note, however, that the results of the simulation are not sensitive to this parameter since it is significantly larger than the later dimension of the photoexcited region (100 μm). The conductivity of metallic K_3C_{60} was set to $8 \times 10^4 \text{ S m}^{-1}$ as measured in previous THz conductivity experiments. The conductivity of the 150 nm aluminum thin film was measured at dc frequency using a van der Pauw configuration in a Quantum Design Physical Property Measurement System. The measurement yielded a conductivity of $\sim 3 \times 10^7 \text{ S m}^{-1}$ at 50 K (the temperature at which we performed the pump-probe experiment; see Fig. 3). We used this measured value in our simulations. We then simulated the expected magnetic field dynamics under two scenarios: (1) assuming a metastable diamagnetic response emerging after photoexcitation and (2) a diamagnetic response persisting only for the duration of the drive. The results of these simulations are presented in Figs. 3(d) and 3(e). The photoexcited area was modeled as a medium that develops a homogeneous, time-dependent magnetic susceptibility $\chi_{\text{photo}}(t)$.

In the first scenario, $\chi_{\text{photo}}(t)$ is modeled as a step function with ~ 1 ps fall time reaching final values of the diamagnetic susceptibility, $\chi_{\text{photo}}(t \rightarrow +\infty)$, equal to the average equilibrium field-cooled magnetic susceptibility of K_3C_{60} at 4 and 16 K [see Fig. 3(c) and Appendix F]. The resulting time traces of $\chi_{\text{photo}}(t)$ are displayed as the blue and red lines in Fig. 3(d), top panel, representing the 4 and 16 K cases, respectively. We performed additional simulations to account for the sample variability assigning to $\chi_{\text{photo}}(t \rightarrow +\infty)$ the maximum and minimum values of the equilibrium diamagnetic susceptibility measured at 4 and 16 K (Appendix F). These values correspond to the limits of the shaded areas in Fig. 3(d), top panel.

The corresponding magnetic field changes are shown in the lower panel of Fig. 3(d). Regardless of the final value $\chi_{\text{photo}}(t \rightarrow +\infty)$, the traces show a long-lived magnetic field change appearing after an initial drop with a time constant of ~ 120 ps. As discussed above, this decay time is due to the inductive response of the unperturbed part of the metallic

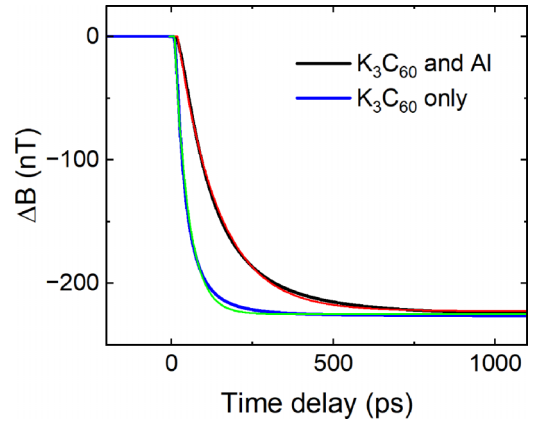


FIG. 12. Simulated magnetic field dynamics for the case of a metastable susceptibility quench with amplitude -0.185 when only the metallic nonexcited K_3C_{60} puck is considered (blue solid line) and when both the puck and the aluminum film are considered (black solid line). The red and green lines are single time-constant exponential decay functions used to fit the data.

K_3C_{60} pellet and the aluminum thin film deposited on the Bi:LIGG detector (see Appendix D). The values of long-lived magnetic field change (ΔB) in the different cases discussed above are summarized in Table I.

In the second scenario, $\chi_{\text{photo}}(t)$ follows a Gaussian pulse peaking at $\chi_{\text{photo}}(t = 0) = \chi_{i, \text{ave}}(4 \text{ K})$ [solid blue line in Fig. 3(e), top panel]. Due to the inductive response of nearby metallic elements, the maximum amplitude of the magnetic field change is severely reduced and is ~ -2 nT, well below our experimental resolution [solid blue line Fig. 3(e), lower panel]. The limits of the shaded blue band in Fig. 3(e) assume $\chi_{\text{photo}}(t \rightarrow +\infty)$ to equal the maximum and minimum measured diamagnetic susceptibilities at equilibrium at 4 K.

Finally, we assessed the impact on the temporal resolution of the experiment of the thin aluminum coating.

The relative contributions to the overall magnetic field shielding of the equilibrium K_3C_{60} sample and aluminum thin film are shown in Fig. 12. This figure shows the simulated results performed under the assumption of a metastable magnetic response of the photoexcited state (see below). The black line represents the case in which the effect of both the K_3C_{60} puck and aluminum film is simulated. The blue line represents the case in which only the K_3C_{60} pellet is accounted for in the model. By fitting the curves with exponentially decaying functions (dashed lines in the same figure), we can extract a time constant of ~ 120 and 40 ps in these two cases discussed. With the aluminum film, the time resolution is worsened by a factor of 3, which does not qualitatively impact the simulated magnetic dynamics discussed above, but the noise of the probe is significantly reduced.

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