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Electron spin resonance insight into broadband absorption of the $Cu_3Bi(SeO_3)_2O_2Br$ metamagnet

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Metamagnets, which exhibit a transition from a low-magnetization to a high-magnetization state induced by the applied magnetic field, have recently been highlighted as promising materials for controllable broadband absorption. Here we show results of a multifrequency electron spin resonance (ESR) investigation of the $Cu_3Bi(SeO_3)_2O_2Br$ planar metamagnet on the kagome lattice. Its mixed antiferromagnetic/ferromagnetic phase is stabilized in a finite range of applied fields around 0.8 T at low temperatures and is characterized by enhanced microwave absorption. The absorption signal is non-resonant and its boundaries correspond to two critical fields that determine the mixed phase. With decreasing temperature these increase like the sublattice magnetization of the antiferromagnetic phase and show no frequency dependence between 100 and 480 GHz. On the contrary, we find that the critical fields depend on the magnetic-field sweeping direction. In particular, the higher critical field, which corresponds to the transition from the mixed to the ferromagnetic phase, shows a pronounced hysteresis effect, while such a hysteresis is absent for the lower critical field. The observed hysteresis is enhanced at lower temperatures, which suggests that thermal fluctuations play an important role in destabilizing the highly absorbing mixed phase. [http://dx.doi.org/10.1063/1.4943534]

I. INTRODUCTION

Broadband absorption of electromagnetic radiation that spans over several decades in frequency is a material's property that is highly praised in modern electronics. Its applications include RF/microwave filtering,[1] optical signal processing,[2,3] electromagnetic interference shielding,[4,5] etc. The common weakness of materials that are used nowadays is a rather narrow absorption range, typically covering only a few decades in frequency.[4] Moreover, the absorption of these materials is in general not controllable via external stimuli. In this respect, the recently discovered controllable broadband absorption in metamagnets is a very promising phenomenon that pledges novel functionality of these materials.[6]

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Metamagnets are magnetic materials that undergo a magnetic-field induced phase transition from a state with low magnetization [typically an antiferromagnetic (AFM) state] into a state with high magnetization [typically a ferromagnetic (FM) state].[7] Because of demagnetization fields, the AFM-FM transition is often more complicated, as an intermediate mixed phase – a phase where both the AFM and FM phases coexist – is stabilized in a finite field range.[7,8] Due to various possible AFM-FM domain configurations[8] a broad excitation spectrum of the mixed phase is anticipated. Indeed, it has been recently demonstrated that in the $Cu_3Bi(SeO_3)_2O_2Br$ planar metamagnet the range of excitations, i.e. absorption, extends over at least ten decades in frequency (from 100 Hz to a few hundreds of GHz).[6] Importantly, as the broadband absorption is limited to the mixed phase of the material, it is thus controllable by the external magnetic field.[6]

The mineral francisite[9] $Cu_3Bi(SeO_3)_2O_2Br$ features two-dimensional pseudo-kagome layers of Cu^{2+} $S = 1/2$ spins.[10] These are coupled by competing nearest-neighbor FM and next-nearest neighbor AFM exchange interactions within the layers, as well as by an additional, much weaker AFM coupling between adjacent layers.[11] In zero magnetic field the compound undergoes Néel ordering at $T_N = 27.4$ K, where a canted, non-collinear ferrimagnetic spin arrangement within individual layers alternates antiferromagnetically between neighboring layers.[11] Such a complicated order is a consequence of competing intralayer exchange interactions and is stabilized by magnetic anisotropy of the Dzyaloshinskii-Moriya type.[12] Magnetic fields perpendicular to the layers that exceed ~0.8 T (at $T \ll T_N$) induce FM order of neighboring layers.[11] In a finite field range around the mean transition field a mixed AFM/FM phase exists.[6] It is stabilized by the demagnetization field of the FM component compensating for any increase of the external field and thus resulting in a constant total internal magnetic field until a homogeneous FM order is established. Therefore, its width is determined by the sample shape and is typically of the order of 0.1 T for plate-like samples.[6] The mixed phase is characterized by strong absorption, as detected through an enhanced imaginary part of the magnetic susceptibility in the kHz range (ac susceptibility measurements) and in the GHz range (electron spin resonance measurement).[6] In order to further characterize the highly absorbing mixed phase, to determine the dependence of the critical fields on the frequency, and to study possible hysteresis effects, we performed a comprehensive electron spin resonance (ESR) investigation.

II. EXPERIMENTAL DETAILS

All measurements were performed on the same single crystal samples as the ones used in our previous study.[6] A typical sample dimension was $3 \times 2 \times 0.2$ mm^3. The samples were grown at 500–550°C by a chemical-transport-reaction method with bromine as the transport agent, using powders prepared by a solid-state reaction.

The ESR experiments were performed on custom-made transmission-type ESR spectrometers[14,15] at the High Magnetic Field Laboratory at the Helmholtz-Zentrum Dresden-Rossendorf, Germany and at the National High Magnetic Field Laboratory, Tallahassee, USA. The investigations were conducted in the temperature range 2.3 – 35 K and frequency range 100 – 480 GHz in the Faraday configuration, with the applied magnetic field perpendicular to the kagome layers. The standard field-modulation technique was used to enhance the signal-to-noise ratio.

III. RESULTS AND DISCUSSION

A typical ESR spectrum recorded in the magnetically ordered state of $Cu_3Bi(SeO_3)_2O_2Br$ is shown in the inset of Fig. 1. It consists of several components. At 240 GHz and 5 K a sharp signal is centered around 0.8 T, while two much broader components are found at 1.8 T and 3.0 T, the latter being by far the most dominant in intensity. With decreasing frequency the two broad components shift to higher fields, while the position of the narrow component remains unchanged (Fig. 1). Clearly, the origin of the narrow and the broad components is different, which is also indicated by the different ESR signal phase of these components (inset in Fig. 1).

The broad ESR components represent FM resonance modes, which were studied before by Wang *et al.*[13] However, their study was limited to frequencies in the range 300 – 490 GHz, therefore, only the

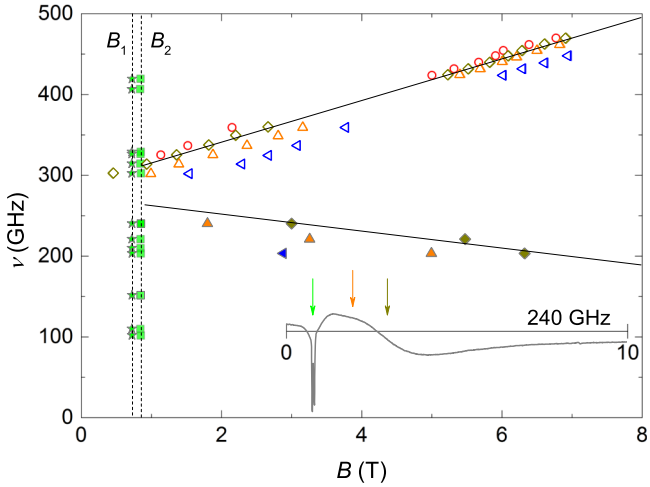

FIG. 1. The frequency-field diagram of $Cu_3Bi(SeO_3)_2O_2Br$ showing various different modes (different symbols). Open symbols represent measurements at 2 K published in Ref. 13, while solid symbols are new measurements performed at 5 K. The vertical dashed lines show the boundaries of the highly absorbing mixed phase characterized by a non-resonant absorption mode. The solid lines go through the positions on the dominant FM resonance mode in both sets of branches. The inset shows the derivative ESR absorption spectrum with three spectral components (arrows), as recorded at 240 GHz and 5 K.

modes where the frequency increases by increasing magnetic field could be detected. Our measurements reveal new FM resonance branches at $\nu < 300$ GHz. At these frequencies we observe one dominant mode and additional weaker modes. This is similar to observations in Ref. 13, where four different branches were observed at $\nu > 300$ GHz, again one being by far the most dominant in intensity. The multiple resonance modes can be due to different sublattices having slightly different exchange anisotropies,[13] or due to surface anisotropy of ferromagnets, which can cause additional modes with slightly different energies.[16]

The main focus of this paper is on the sharp ESR component, which represents a non-resonant absorption mode. There is no dependence of this mode on frequency, as found in the entire frequency range (100 – 480 GHz) of our investigation (Figs. 2 and 3). The critical fields B_1 and B_2, which we define as the lower and the higher borders of the absorption signal, respectively, exhibit temperature dependence resembling that of an order parameter of the AFM phase (Fig. 3) and coincide with the boundaries of the mixed AFM/FM phase of the investigated compound.[6] The

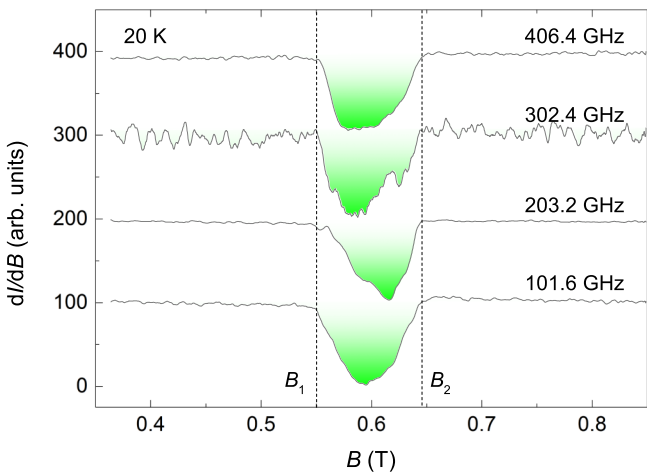

FIG. 2. A collection of non-resonant absorption spectra recorded at 20 K and at various frequencies. Each spectrum is offset vertically by the valued of the corresponding frequency. The vertical lines show the lower B_1 and the upper B_2 critical fields, corresponding the the boundaries of the highly absorbing mixed phase in $Cu_3Bi(SeO_3)_2O_2Br$.

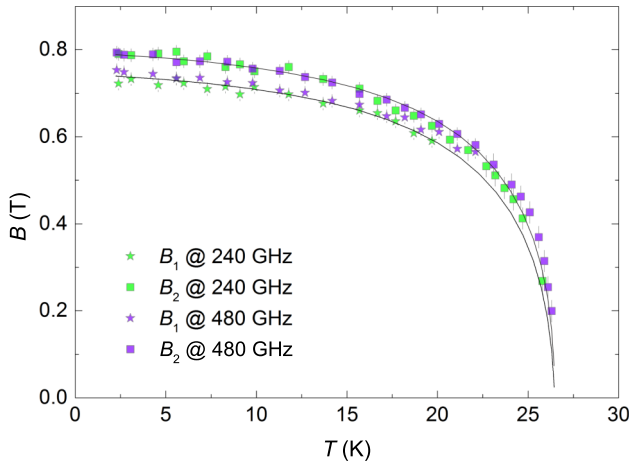

FIG. 3. The temperature dependence of the lower B_1 and the upper B_2 critical fields in $Cu_3Bi(SeO_3)_2O_2Br$ at two selected frequencies behaving like an order parameter of the AFM phase. The solid lines are a guide to the eye.

absorption properties of this phase are notably different from the properties of the two neighboring homogeneous phases and results in the observed non-resonant mode.

In order to better characterize the transition from the AFM into the mixed phase at B_1 and from the mixed into the FM phase at B_2 we measured the non-resonant absorption signal upon sweeping the magnetic field up and down. A pronounced hysteresis is found in the value of the higher critical field B_2, which depends significantly on the direction of the field sweep (Fig. 4). Moreover, this hysteresis effect is strongly temperature dependent. It corresponds to a 2.0% increase of B_2 in the sweep-up experiment compared to the sweep-down experiment at 20 K, while this difference increases to 5.5% at 5 K (Fig. 5). At this temperature the total field width of the mixed phase is as much as 30% larger when increasing the field than when decreasing it. In contrast, within experimental errorbars there is no hysteresis observed in the lower critical field B_1 (Fig. 4) at either of the two temperatures.

Our ESR investigation of $Cu_3Bi(SeO_3)_2O_2Br$ thus confirms that the first-order transition from the low-field AFM phase into the high-field FM state is spread over a range of applied fields, which defines the mixed AFM/FM phase that is highly absorptive. Both boundaries shows an AFM order-parameter-like temperature dependence. Any hysteresis effects are absent in the value of B_1, as expected on the border between the AFM phase and the mixed phase with the FM-phase fraction

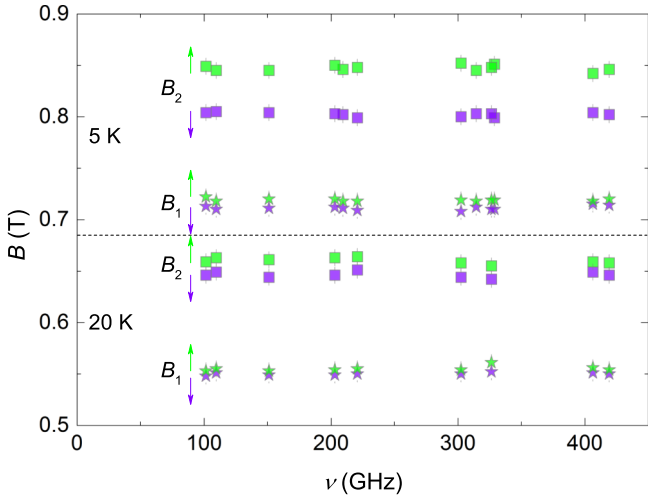

FIG. 4. The frequency dependence of both critical fields at two temperatures (separated by the dashed line). The direction of the field sweep in the experiments is indicated by vertical arrows.

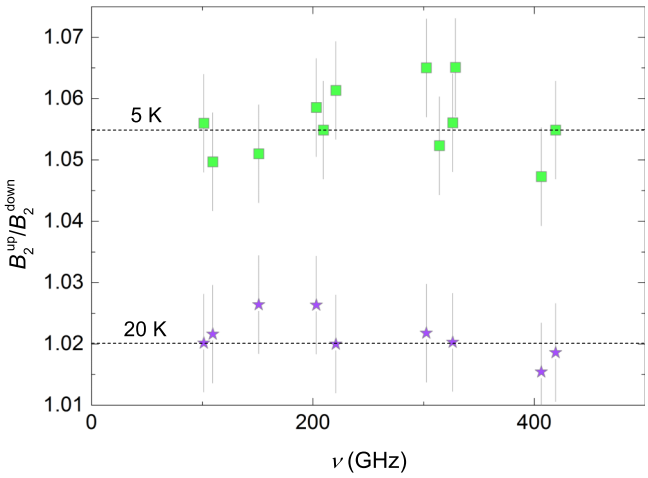

FIG. 5. The hysteresis of the upper critical field between the mixed and the ferromagnetic phase for experimental field sweep up and down at two selected temperatures. The horizontal lines indicate the average value.

being zero at this field. On the contrary, the transition from the mixed phase into the FM phase at the critical field B_2 shows a hysteresis. Its presence is unlikely related to usual mechanisms encountered in FM materials, where the hysteresis corresponds to changing domain structure in multiple-domain ferromagnets or reorientation effects in single-domain ferromagnets. Moreover, the width of the hysteresis is not proportional to the value of the magnetization in the FM phase,[6] as the former increases with decreasing temperature much more profoundly. This experimental observation suggests that thermal fluctuations play an important role in destabilizing the mixed phase. At higher temperatures, where these are more pronounced, the mixed phase is less stable to increasing magnetic field, the history effect becomes less important and, consequently, the hysteresis in B_2 tends to decrease.

IV. CONCLUSIONS

In summary, we performed a systematic ESR study of the $Cu_3Bi(SeO_3)_2O_2Br$ metamagnet, focusing on the non-resonant absorption that is associated with the mixed AFM/FM phase of this material. Our multifrequency experiments have shown that the enhanced absorption in this phase is frequency independent in the entire region of our investigation (100 - 480 GHz). Moreover, we found that the phase boundary between the high-field FM phase and the mixed phase shows a hysteresis effect, while the boundary between the mixed and the low-field AFM phase does not. The hysteresis is suppressed by temperature, implying that thermal fluctuations may play a role in effectively destabilizing the mixed phase. This should be considered in any future modeling of the mixed phase of $Cu_3Bi(SeO_3)_2O_2Br$ and thus in the ultimate search of a microscopic mechanism of enhanced absorption of metamagnets.

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