Coherent Inelastic Light Scattering from a Microwave-Excited Array of Magnetic Particles

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Inelastic light scattering from an array of Permalloy particles driven by a microwave magnetic field is shown to be a coherent phenomenon in which the scattered radiation is observed only at diffraction angles corresponding to the reciprocal lattice of the array. The results are explained in terms of the phase coherence of the inelastically scattered light by each of the particles.

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Coherent scattering of radiation is responsible for all forms of diffraction of electromagnetic waves. Diffraction occurs when radiation, originating from different spatial locations, interferes constructively in the far field. A common example of coherent scattering is found in x-ray diffraction, but it also occurs for visible radiation impinging on samples patterned at length scales comparable to the wavelength of the radiation. The most common example of this latter situation is the diffraction by gratings, which is extensively used in spectroscopy applications. More recently the magnetic field dependence of light diffracted by arrays of patterned magnetic materials has been exploited to extract information on magnetization reversal mechanisms [1,2].

The examples in the preceding paragraph are all related to elastic scattering—where the frequency of the scattered light is unchanged from that of the incident radiation. Here we investigate the phenomenon of inelastic scattering of visible light by an array of magnetic nanoparticles excited by an external oscillating field. Using Brillouin scattering we show that the inelastically scattered light is also diffracted and that it is confined to the same diffraction directions as the elastically scattered light.

Brillouin scattering is extensively used to study spin excitations in magnetic systems including nanoarrays [3–6]. In these experiments the thermally populated excitations that are observed correspond to the low-lying spin modes that lie close to, and may include, the uniform ferromagnetic resonance mode (FMR) of each particle in the array. Brillouin scattering has also been used to study excitations in magnetic thin films driven by an external microwave field [7–14]. These latter studies have dealt primarily with the interpretation of the magnetic excitations and not with the scattering aspects of the phenomenon. Here we wish to highlight a rather unexpected aspect of the light scattering process itself when it is used to investigate driven excitations in an array of magnetic particles.

We will show that inelastically scattered light from non-driven magnetic nanoarrays is not diffracted because the excitations in each particle are not phase coherent. The resulting scattered light thus shows no strong angular dependence. We will also show that the inelastically scattered light from a *uniformly driven* array of magnetic particles is diffracted in the same way as the elastic scattering. Inelastically scattered light is observed only along narrow, highly collimated directions in space. The coherent nature of the externally driven excitations explains this diffraction effect.

The setup used in our experiments is shown schematically in Fig. 1(a). The array of short magnetic bars, deposited on a 20 μm thick Si substrate, is placed on a 100 μm wide microwave stripline. A thin substrate (less than the width of the stripline) is necessary in order to keep the

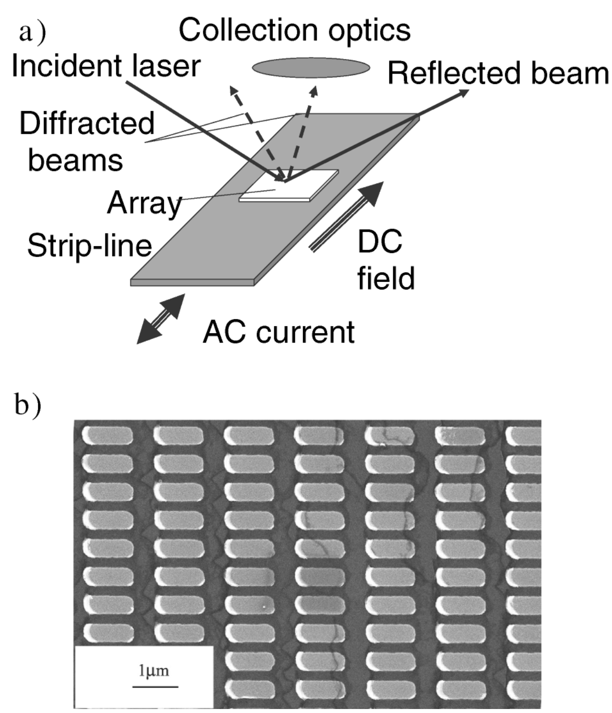

FIG. 1. (a) Schematic of experimental setup. (b) SEM image of array.

array within the region with the highest fields produced by the stripline. Figure 1(b) is the scanning electron microscope (SEM) image of the array of 20 nm thick Permalloy elongated particles. The microstripline of 50 Ω characteristic impedance is in turn placed between the poles of an electromagnet with the current and the long axis of the particles both parallel to the applied static magnetic field. When the full power of our microwave generator is applied to the microstripline the current produces an oscillating field of about 0.7 Oe in the plane of the sample and perpendicular to the static magnetic field. The whole setup is mounted in a manner that permits Brillouin spectra to be recorded. Figure 1(a) also shows the incident and scattered laser beams and two of the diffracted beams. One of the diffracted beams enters the collection optics for the Brillouin experiments. Our spectra were recorded using 200 mW of 515 nm radiation from a single-mode Ar laser. Spectra were recorded in both 3 + 4 tandem and 5-pass nontandem, operation of a Sandercock interferometer [15]. The latter configuration yields higher count rates, the former enables evaluation of possible effects due to overlapping orders. Because of the large and well known Stokes–anti-Stokes (S, AS) intensity asymmetries that exist for magnetic systems [16], we find that there is negligible overlap between Stokes and anti-Stokes portions of the spectra.

Before describing the spectra we note that the diffracted beams originating from the patterned sample are clearly visible to the naked eye. One of them enters the collection lens aperture. This beam can be identified as the (0, 1) beam lying in the scattering plane. The diameter of the diffracted beam at the collection lens is around 2 mm.

In Fig. 2 we show Brillouin spectra recorded under different experimental conditions. Spectrum (a) was acquired with zero applied microwave driving power and a large collection aperture ($f/3.5$) with the sample in a 1 kOe field. It is a typical Brillouin spectrum and the two peaks correspond to two thermally excited modes at 10.4 and 12.1 GHz. The spectrum in (b) is the same as in (a) except that the collection angle has been substantially narrowed so that the solid angle collected is reduced by about a factor of almost 100. The amount of inelastic light seen in the Brillouin spectrum is also substantially reduced compared to (a), showing that in the nondriven system the inelastic light is scattered nearly isotropically.

The results in Figs. 2(c) and 2(d) are dramatically different. Spectrum (c) was recorded by applying microwave power to the stripline at a frequency of 10.4 GHz and with the large aperture. Clearly there is a significant enhancement to the inelastic scattering from the mode at 10.4 GHz. Since the Brillouin signal from a magnetic excitation scales as the square of the precession amplitude, the enhanced signal can be understood as due to an increase in amplitude caused by the driving field. Spectrum (d) in Fig. 2 was obtained under identical conditions as spectrum (c) but the collection aperture was occluded to a small opening allowing only the visible (0, 1) diffracted peak to be collected. Again this corresponds to a reduction of almost 100 in the collection solid angle, so it is amazing that no appreciable intensity is lost from the uniform mode. (The observed increase is within the uncertainty of the overall alignment errors of the optics.) Thus we have shown that *all* the inelastically scattered light is limited to the directions of the static diffraction when the sample is

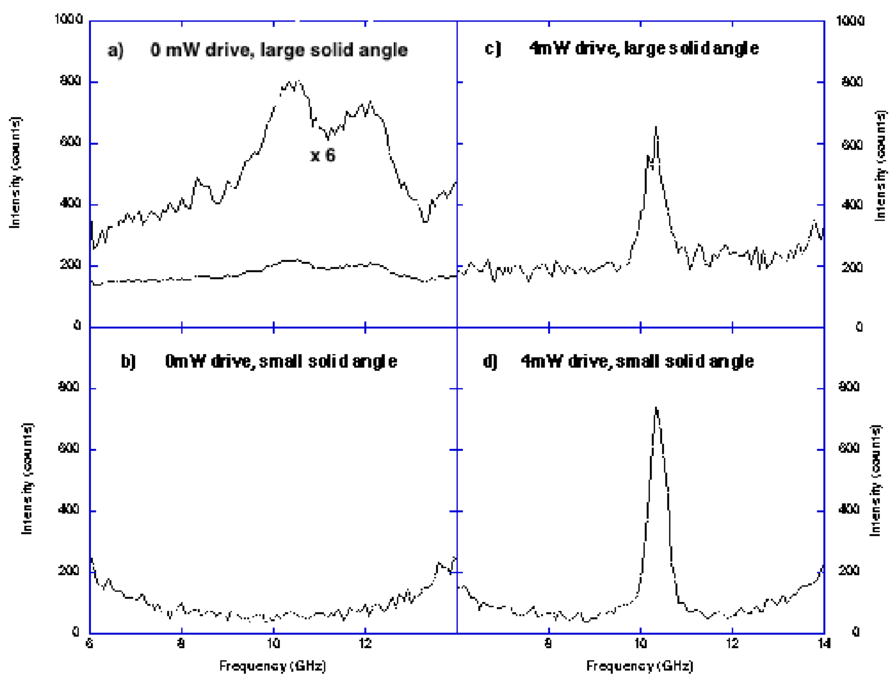

FIG. 2 (color online). Brillouin spectra vs power and solid angle at 1 KOe. The upper line in (a) shows the lower line multiplied by a factor of 6 and offset.

driven by a uniform microwave field. Hence we label it "coherent inelastic scattering."

Another feature exhibited by this coherent scattering is its "resonancelike" behavior. In Fig. 3 we plot the intensity of the enhanced Brillouin peak [as seen in Fig. 2(d)] as a function of the driving frequency for three values of the applied static magnetic field. Clearly coherent scattering occurs only when the driving frequency is within the resonance band of the spin mode. We have also ascertained that the intensity of the Brillouin peak scales with the driving power. We found no resonance enhancement of the 12.1 GHz mode, observed at 1 kOe in Fig. 2(a), even when the system was driven at 12.1 GHz.

We now turn to an explanation of this behavior. Elastic scattering (the static diffraction pattern) occurs where the EM radiation, induced by the incident wave, emanating from each bar interferes constructively. In contrast, inelastic scattering is caused by electromagnetic waves interacting with thermal magnetic excitations already present in each of the magnetic bars. With no driving field, the phase of the spin waves in each particle is random and the inelastic scattering is incoherent with no preferential directional dependence in the inelastically scattered light. Under this condition the intensity should be proportional to the collection solid angle as is seen in Figs. 2(a) and 2(b). However, when the driving field synchronizes the precession of the spins in all the particles, one creates the same conditions as in the static diffraction: destructive interference occurs for all scattering directions except along the static diffracted beams where all contributions are in phase; see Figs. 2(b) and 2(d).

To understand the details of the magnetic scattering, we calculated the normal modes of a $1120 \times 360 \times 20\ \mathrm{nm}^3$ Permalloy particle using the technique described in [17]. From these calculations we can identify the two peaks seen in Fig. 2(a). The lower frequency peak at 10.4 GHz can be

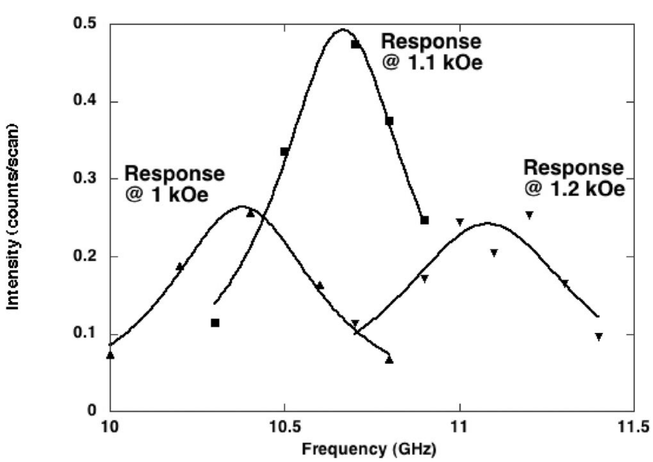

FIG. 3. Resonance behavior of the enhanced excitation at various applied fields. The symbols are the measured Brillouin intensities for three different applied fields. The solid lines are fits to Lorentzian line shapes.

identified as the uniform precession mode (equivalent to the FMR mode with the appropriate shape anisotropy fields). The higher frequency mode at 12.1 GHz is a mode which is odd along the long axis of the bar. These two modes were identified since they would be expected, based on wave vector considerations, to produce strong scattering in a Brillouin experiment. The profiles of these modes are shown in Figs. 4(a) and 4(b).

One can now understand why the low frequency mode is the only mode that occurs in the driven spectrum. One can see from Fig. 4 that the uniform mode [Fig. 4(a)] will couple to a uniform driving field, while the odd mode at 12.4 GHz will not be excited. This explains why the mode at 12.4 GHz does not exhibit coherent scattering even if it is driven with a 12.4 GHz microwave field. We speculate that this mechanism might operate as a filter for identifying spin wave modes: only modes which have significant "overlap" with the uniform mode can show up in the driven Brillouin spectrum.

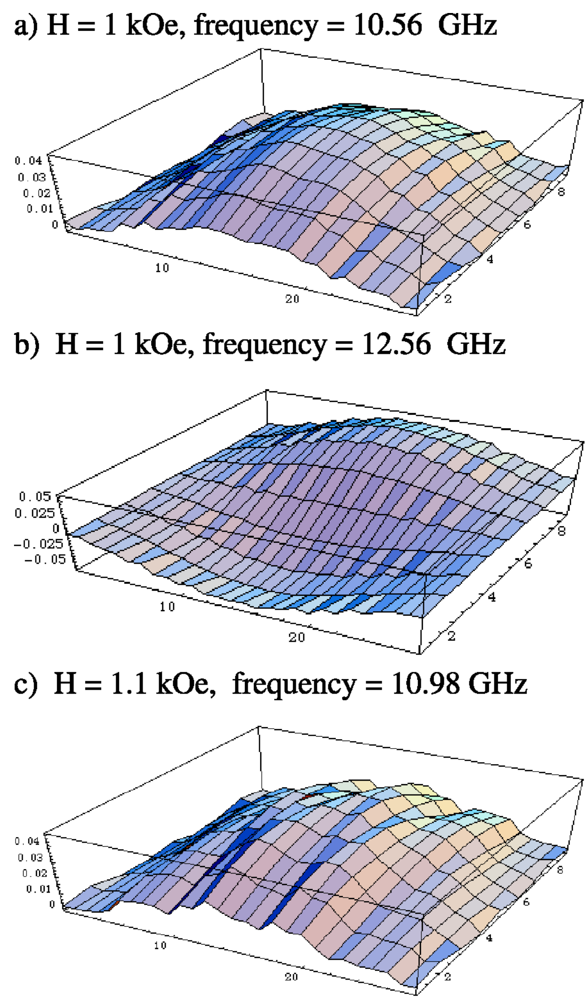

FIG. 4 (color online). Profiles and frequencies of the spin excitations at two fields. (a) and (b) correspond to the modes observed in Fig. 2(a). The profile in (c) shows the effect of hybridization on the fundamental mode.

The explanation given in the above paragraph provides a simple physical picture for the origin of diffraction of inelastically scattered light. The process can also be described more formally by treating each particle as a unit cell of a crystal. In this formalism the excitation driven by the microwave field is a zone center mode of the array; i.e., all unit cells in the crystal oscillate in phase. In this picture the scattering process is an Umklapp process in which a zone center ($q = 0$) excitation is observed at a reciprocal lattice vector G. In this case $G = 2\pi(0, 1)/a$ where a is the lattice constant of the array.

We have also verified experimentally that the coherent scattering obeys the same selection rules encountered in Brillouin scattering from thermally excited spin excitations. Resonant magnon peaks are obtained only in cross polarizations of the incident and scattered light (i.e., only for s-p or p-s polarizations). The S:AS intensity ratio reverses on reversal of the magnetic field, and the ratio of the S and AS intensities is different for s-p and p-s polarizations.

The above discussion accounts for all effects highlighted in Fig. 2 and qualitatively for the resonancelike behavior, at any given field, in Fig. 3. In a forced oscillatorlike model, if the driving frequency lies outside the bandwidth of the mode, driving will not occur and the effect will vanish as shown in Fig. 3. What is less clear is why the width of the resonance curves in Fig. 3 should be so strongly field dependent. If we view the width and/or height of the resonance curves as a measure of the excitation lifetime, why should it change so drastically, and nonmonotonically, with applied field? We believe that this effect could be due to hybridization between the uniform resonance mode and other normal modes of the particle. In our calculations of the normal modes we detect numerous mode crossings as the field is changed. At some of these crossings considerable hybridization occurs as can be seen in Fig. 4(c) where we show that profile of the ''uniform'' mode at 1.1 kOe (identified by its frequency and its nonzero value of the spatial average of its amplitude). Hybridization is evidenced by the additional wiggles superimposed on the structure shown in Fig. 4(a). At a field of 1.13 kOe the mixing of the modes is so large that the uniform mode becomes almost unrecognizable. Such hybridization effects have also been observed in simulations of the normal modes of similarly shaped particles [17]. These near degeneracies that occur for certain field values may lead to a reduced lifetime of the FMR mode and thus explain the observed broadening of the resonance. This aspect of the problem clearly deserves a more careful investigation and will be pursued in future experiments.

We have shown the existence of the phenomenon of coherent inelastic scattering. This phenomenon may prove to be a useful tool for investigating nonlinear magnetic phenomena. We envision that with larger amplitude microwave driving fields, that can be achieved both by higher power and by reducing the sample to microstripline distance, it should be possible to drive specific spin modes into their nonlinear regimes. This would provide new data on magnetic nonlinearities, information on magnetic damping effects, and may yield information on the interaction between spin excitations. This latter aspect could be of great importance for technological applications of magnetic nanoparticles. Preliminary studies with microwave fields 20 times larger than those used in the present experiments yield very complex behavior, including frequency shifts of the main resonance line, indicating the nonharmonic nature of the resonance at these fields.

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