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Intrinsic ferrimagnetic resonance linewidth of barium ferrite due to spin-wave scattering by trigonal site single-particle excitations

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Time-dependent two-magnon scattering was previously proposed as a mechanism to explain the large magnitude of the ferrimagnetic resonance (FMR) linewidth of barium ferrite as a function of frequency. In the present work, it is shown that a quantum mechanical mechanism like the Kasuya–Le Craw process (KL)¹ but with the phonon excitation replaced by a single-particle excitation of a trigonal site iron ion, which moves in an anharmonic potential well, gives a linewidth contribution of less than a tenth of an Oersted and proportional to the frequency, as in the KL mechanism. We conclude, based on this work and our previous work on the KL mechanism, that neither of these mechanisms can explain the observed FMR linewidths in barium ferrite at any frequency.

I. INTRODUCTION

Much of the previous work on ferrimagnetic resonance (FMR) relaxation is based on the work of Kasuya–LeCraw (KL).¹ Most of the details of that work are published in the book by Sparks.² The KL mechanism predicts an FMR linewidth that is linear in the FMR resonance frequency and nearly linear in the temperature.² Measurements on high-quality spherical and planar single-crystal samples (as determined from the low value of the FMR linewidth and strikingly large temperature dependence down to low temperatures) give a linewidth of magnitude 30 Oe between 50 and 64 GHz, nearly linear with temperature.³ Measurements made on the spherical samples over FMR resonant frequencies ranging from about 40 to 105 GHz, however, are more consistent with a linewidth that is nearly independent of resonant frequency and approximately equal to 80 MHz or 30 Oe.⁴ Tsantes and Silber⁵ proposed that the high intrinsic linewidth of barium ferrite could be due to a dynamical two magnon process in which the uniform magnon is scattered into higher frequency modes by random time fluctuations of the trigonal site ion, which is believed to hop at temperatures greater than 77 K between two equilibrium sites 0.16 Å to either side of the symmetry plane.^{6–10} This process was argued to give a linewidth independent of resonant frequency.⁴ The magnitude of the contribution from this process and from the KL mechanism to the FMR linewidth has been estimated for barium ferrite¹¹ by us using detailed calculations of the magnon and phonon dispersion relations.^{12,13} The estimate for the dynamical two magnon process was about 15 Oe at about 100 GHz at room temperature, whereas even the most optimistic estimates for the contribution from the KL mechanism only gave a value of about 1 Oe, consistent with the former being the dominant mechanism.

The lattice dynamical studies of Ref. 13 were shown to be consistent with soft mode behavior for a mode that consists primarily of trigonal site iron motion relative to its neighboring oxygens, which is expected if the trigonal site iron ion either lies in a fairly flat-bottomed potential well or if there is a phase transition at a certain temperature at

which the double well changes over to a single well. This implies that the dynamics of this ion might be better described by motion of a single particle in a flat potential well with steep sides (to represent the hard core repulsion by the neighboring oxygen ions) or such a well with a low barrier in the middle (i.e., a double potential with a weak barrier in the middle) than a well whose minimum lies at the symmetry plane.

The linear dependence of the linewidth on FMR resonant frequency and nearly linear dependence on temperature, which is characteristic of the KL mechanism, comes about because of the KL population factor

$$n(\omega_{sw})n(\omega_{ph})\hbar\omega_u/k_B T,$$

where $n(\omega)$ is the Bose–Einstein-function, $(e^{\hbar\omega/k_B T} - 1)^{-1}$, ω_{sw} and ω_{ph} are the spinwave and phonon frequencies, and ω_u is the uniform spinwave or FMR resonant frequency. This expression was obtained using somewhat heuristic arguments on pp. 93 and 94 of Ref. 2, and it was also obtained more rigorously using a many-body perturbation theory for the ac susceptibility in Ref. 14 in an Appendix. It is quite likely that the main reason that the KL mechanism's contribution to the FMR linewidth is so much smaller than the dynamical two magnon scattering mechanism of Ref. 5 is that the former process contains a factor $\hbar\omega_u/k_B T$, whereas the latter does not.^{5,11} The latter assertion will be challenged in the present article.

The calculations of Ref. 11 give the linewidth due to the dynamical two magnon scattering in terms of the frequency of the trigonal site ion oscillatory motion, but Ref. 11 does not discuss how this frequency is related to the temperature in any detail. In the present work, a quantum mechanical mechanism, like the KL process but with the phonon excitation replaced by a single-particle excitation of a trigonal site iron ion, which moves in an anharmonic potential well, gives a linewidth contribution of less than a tenth of an Oersted and proportional to the frequency, as in the KL mechanism.

II. CONSEQUENCES OF THE SINGLE-PARTICLE MODEL FOR TRIGONAL SITE MOTION

A good model for the contribution to the FMR linewidth due to the anharmonic motion of the trigonal site ion is to treat this ion as a particle in a one-dimensional potential well. The linewidth results from the uniform magnon scattering into a magnon of energy that is higher than that of the uniform magnon by the energy difference between two trigonal site ion single-particle energy levels. In other words, the creation or annihilation of a phonon in the KL mechanism is replaced by a single-particle excitation of the trigonal site ion in its potential well. Since the trigonal site single-particle states are essentially localized states, the spinwave wavevector is not conserved (just as occurs for the time fluctuating two magnon scattering process^{5,11}). If the potential in which the trigonal site ion moves is harmonic, this reduces to a KL mechanism. In general, the expression for the linewidth is equal to the difference between the process in which the ion moves down (resulting in the creation of a spinwave whose energy is higher than the uniform spinwave by the difference between the single-particle energy levels) and one in which the ion moves up in energy, averaged over initial states of the trigonal site ions and the magnons. The linewidth is thus proportional to

$$Z^{-1} \sum_{n_1=1}^{\infty} \sum_{n_2=n_1}^{\infty} \{ |M_{n_2, n_1}|^2 e^{-E_{n_2}/k_B T} [n(\omega_{sw}) + 1] - |M_{n_1, n_2}|^2 e^{-E_{n_1}/k_B T} n(\omega_{sw}) \} \rho(\omega_{sw}), \quad (1)$$

where E_n is the energy of the trigonal site ion in the n th energy level, $Z = \sum_n e^{-E_n/k_B T}$, M_{n_2, n_1} is the matrix element of the trigonal site ion single-ion anisotropy, which causes the single-particle excitation, and $\rho(\omega)$ is the linewidth due to the periodically oscillating ion calculated in Ref. 11 divided by ΔD^2 (where ΔD is the difference between the value of the single-ion anisotropy when the ion is on the symmetry plane and when it is 0.16 Å to one side of the plane). The population factor for the spinwaves is $n_{sw} = n(\omega_{sw})$, where $\omega_{sw} = \omega_u + [(E_{n_2} - E_{n_1})/\hbar]$ if the ion moves up in energy (resulting in the absorption of a spinwave whose energy is higher than the uniform spinwave by the difference between the single-particle energy levels) and $n(\omega_{sw}) + 1$ if the ion moves down in energy. (It should be noted that for the special case where the trigonal site ion moves in a harmonic oscillator potential, n_2 and n_1 can only differ by 1.) Equation (1) may be rewritten as

$$Z^{-1} \sum_{n_1=1}^{\infty} \sum_{n_2=n_1}^{\infty} |M_{n_2, n_1}|^2 e^{-E_{n_1}/k_B T} n(\omega_{sw}) \times [e^{\{\hbar\omega_{sw} - (E_{n_2} - E_{n_1})/k_B T\}} - 1] \rho(\omega_{sw}). \quad (2)$$

From the definition of ω_{sw} given above we see that the factor in square brackets reduces to

$$e^{\hbar\omega_u/k_B T} - 1. \quad (3)$$

Since at room temperature $\hbar\omega_u \ll k_B T$, we see that the FMR linewidth is always proportional to ω_u since none of the

other population factors have a significant dependence on ω_u . This is because all of the other population factors that depend on it are functions of the sum or difference between it and the single-particle energy spacing, which is much larger in comparison. The only time it is correct to treat the trigonal site ion as a classical harmonically oscillating scattering potential, as was done in Ref. 11, is in the limit in which the trigonal site level spacing $E_{n_2} - E_{n_1}$ is small compared to the temperature and to all the spinwave energies in the problem. In that limit, if the trigonal site potential were harmonic, we would obtain a linewidth independent of FMR resonant frequency. This is because in that limit the thermal population factor for the KL process given in the introduction contains a factor of ω_u in its denominator that cancels out ω_u in the numerator. For the case of a harmonic potential, $E_{n+1} - E_n$ is independent of n and is equal to \hbar times the oscillator frequency. The matrix element M_{n_2, n_1} is proportional to $\delta_{n_2, n_1} (n_1 + 1)^{0.5}$ (Ref. 15) and these factors give the usual KL population factors, since the average of n is the usual Bose-Einstein function of the harmonic oscillator frequency, as occurs in the KL mechanism. The population factor in the KL mechanism given at the end of the last section is the difference between the phonon and magnon Bose-Einstein functions. For most nonharmonic potentials, the matrix element of the ion displacement does not in general have this form, as we shall see. For example, consider a one-dimensional infinitely deep square well potential of width L as a model potential for the trigonal ion. If the origin is placed at the center of the well, the wave function is

$$(2/L)^{0.5} \cos(n\pi x/L), \quad (4a)$$

if the integer n is an odd integer, and

$$(2/L)^{0.5} \sin(n\pi x/L), \quad (4b)$$

if n is even and energy levels given by $(\hbar^2 \pi^2 n^2)/(2mL^2)$, where m is the ion mass. Then the absolute value of the matrix element of x between the states with $n = n_1$ and n_2 is easily calculated to be

$$[(-1)^{(n_1+n_2+1)/2} (n_2 - n_1)^{-2} + (-1)^{(n_1+n_2-1)/2} (n_1 + n_2)^{-2}] L/\pi, \quad (5)$$

whose thermal average will not be a Bose-Einstein function of the level spacing, as it is for a harmonic oscillator. The matrix element M_{n_1, n_2} is equal to Eq. (5) multiplied by the derivative of the anisotropy coefficient with respect to the displacement of the trigonal site ion. This is to a good approximation Eq. (5) with L replaced by ΔD , defined under Eq. (1) since L represents the ion displacement and ΔD is to a good approximation the derivative of D multiplied by L . Using this expression for M_{n_1, n_2} and ρ from Ref. 11, we have integrated Eq. (1) numerically to find the contribution of this mechanism to the linewidth. The results are given in Fig. 1 for $\omega_u = 100$ GHz. The change in the single ion anisotropy used is the value 0.5 cm^{-1} used in Refs. 5 and 11. Although the linewidth increases with increasing temperature, as is observed for pure barium ferrite single crystals, the magnitude of the contribution is only of

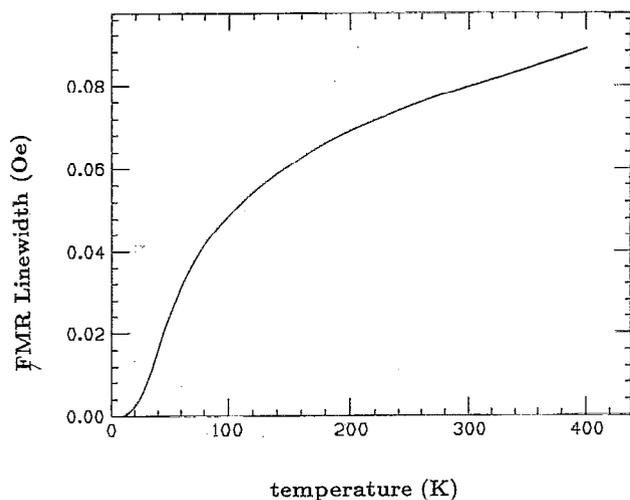


FIG. 1. FMR linewidth at a resonant frequency of 100 GHz for the single-particle model calculated from Eq. (1) in Oe as a function of temperature in K.

the order of a tenth of an Oe, which is comparable to the linewidth estimates for YIG done by Sparks. The primary reason for this is that this mechanism contains a factor $\hbar\omega_n/k_B T$, which was thought originally not to occur in the expression for the linewidth contribution due to the fluctuating ion mechanism.¹¹ As a result of the presence of this factor the results found in Ref. 11 are reduced from about 15 Oe to at most a tenth of an Oe. The case of a barrier in the middle of the trigonal site ion potential well can be treated semiquantitatively by assuming that the lowest single-particle mode occurs at an energy above the assumed barrier energy and then increases with increasing n as E_n given above does. This neglects the small contribution from the levels that lie below the barrier for which the quantum mechanical tunneling motion is assumed to be small.

III. CONCLUSIONS

We concluded in this article that the fluctuating ion mechanism (which is only able to give an intrinsic linewidth of at most 1 Oe) is not able to explain the observed magnitude of the FMR linewidth of barium ferrite. We concluded in Ref. 11 that the KL mechanism was also unable to account for the smallest linewidths observed for barium ferrite of about 30 Oe. Since neither of these mechanisms are able to explain the measured linewidths, the possibility exists that the intrinsic linewidth limit has not yet been achieved in measurements on barium ferrite. Other possible mechanisms will be considered in future work.

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