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LETTER TO THE EDITOR

Evidence for domain wall tunnelling in a quasi-one dimensional ferromagnet

Kimin Hong and N Giordano

Department of Physics, 1396 Physics Building, Purdue University, West Lafayette, IN 47907-1396, USA

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Abstract. We have studied the motion of magnetic domain walls in very small diameter ferromagnetic wires. Our results suggest that walls do not move smoothly along the wire, but can be trapped at pinning sites. It appears that they escape from these sites by thermal activation at high temperatures, and quantum tunnelling at low temperatures. Tunnelling dominates below $\sim$ 5 K, which is about an order of magnitude higher than predicted by the current theory.

In recent years, microfabrication methods have improved significantly, leading to new opportunities with regard to fundamental physics. One problem in this area which is of particular interest concerns the quantum behaviour of 'macroscopic' objects. Leggett and coworkers [1] have emphasized the importance of such macroscopic quantum phenomena (MQP) in tests of quantum mechanics. To date, the most detailed experiments have involved Josephson junctions [2] (although there is also evidence in other systems [3]), where it has been possible to observe tunnelling of the collective coordinate which describes the superconducting phase difference across the junction, an effect known as macroscopic quantum tunnelling (MQT). Several years ago it was proposed [4–6] that MQP should be observable in magnetic systems, involving small particles or domain walls. While there appears to be reasonable experimental evidence supporting this view [6, 7], a difficulty with the experiments is that they essentially all involve collections (i.e. ensembles) of tunnelling entities [8]. The fact that tunnelling rates vary exponentially with parameters such as system size, together with the inevitable variations of these parameters, complicates the interpretation of the experiments. It would clearly be desirable to study the behaviour of a *single* tunnelling entity. In this letter we describe an experiment which may have accomplished this, and present evidence for the motion of individual domain walls via MQT.

The samples were small diameter ferromagnetic wires fabricated from evaporated Ni films, using a step-edge technique [9]. Typical wire diameters were 200–400 Å, with lengths between Cu contact leads of $\sim$ 10 μm (the Ni wires extended for a much greater distance beyond the contacts). The low temperature (residual) resistivity of the wires was $\sim$ 10 $\mu\Omega$ cm, which implies that the elastic mean free path was limited by boundary scattering. The grain size of these polycrystalline Ni films was $\sim$ 25 Å, much smaller than the wire diameters. The resistance was measured using standard ac bridge techniques, with a magnetic field applied parallel to the axis of the wire [10].

Typical results for the resistivity, ρ, as a function of field are shown in figure 1. In both cases the field was swept at a constant rate ($\sim$ 0.5 Oe s^{-1}). The solid curve was obtained while sweeping the field down, while the dotted curve was measured while sweeping up,

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just after exposing the sample to a large negative field. The difference between the two results was due to differences in the sample magnetization. The large positive field applied prior to measuring the solid curve caused the sample to be uniformly magnetized in the positive direction; thus there were no domain walls in the sample. The solid curve was completely reversible and reproducible; the sweep could be interrupted or reversed, with no change in the $\rho(H)$ relation. The large negative field applied prior to measuring the dotted curve caused the sample to again be uniformly magnetized, but this time $\boldsymbol{M}$ was in the negative direction. The dotted curve (including its extension to negative fields, which is not shown here) was also completely reversible, so long as the field was not swept past the minimum point of the curve. However, once the field passed this point, the behaviour was hysteretic.

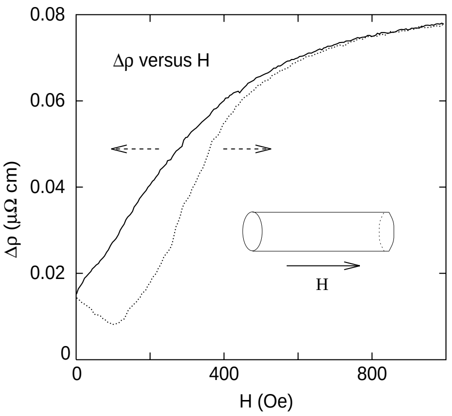

Figure 1. Resistivity of a 300 Å diameter Ni wire as a function of magnetic field at 4.2 K. The solid curve shows the behaviour when sweeping the field down from large positive values (as indicated by the dashed arrow); the dotted curve was obtained while sweeping H up (also indicated by the dashed arrow), after coming from a large negative field. (Note that the zero of the vertical scale is arbitrary, since we are plotting only the change in ρ.)

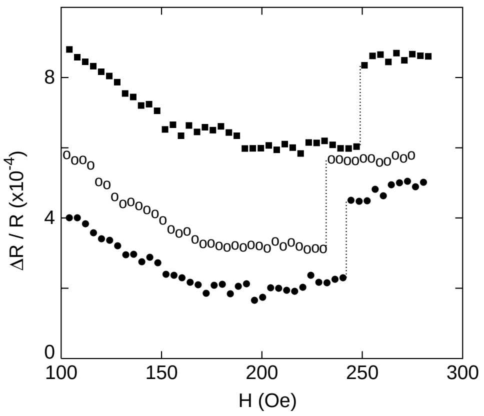

Figure 2. Repeated measurement of the resistance of a 300 Å Ni wire. The vertical dotted lines indicate the jumps discussed in the text. The field was swept at 0.5 Oe s^{-1}. For clarity, the different data sets have been offset vertically.

These results indicate that, when following the dotted curve in figure 1, the sample did not contain any domain walls until the field was increased to about 150 Oe. Above this field domain walls must have entered the wire, so that different portions of the sample then had $\boldsymbol{M}$ in different directions. The sample shape and size make this magnetization reversal process particularly simple. First, at these fields demagnetizing effects (i.e. shape anisotropy) cause the magnetization to lie along the axis of the wire. Hence, there are only two possible directions for $\boldsymbol{M}$, which we have already loosely referred to as positive and negative. Second, the domain walls should have a thickness [11] of at least $\sim$ 1000 Å. Since this is much greater than the wire diameter, the walls should, to a first approximation, simply slice through the wire, with their normals parallel to the wire axis. Walls which meander through the wire, or which run along the axis, would not be favored, since they would cost too much energy. When moving along the dotted curve in figure 1, a sufficiently large field caused domain walls to enter the sample at $H \sim 150$ Oe [13, 14]. These walls separated regions in which $\boldsymbol{M}$ is parallel or antiparallel to the axis of the wire. As the reversal process proceeded, walls moved along the wire, as domains with $\boldsymbol{M} \parallel \boldsymbol{H}$ grew at the expense of those with $\boldsymbol{M}$ in the opposite direction.

The difference between ρ at the minimum of the dotted curve in figure 1, and the value at the same field for the solid curve is a type of longitudinal magnetoresistance [15]. It depends on the relative orientation of $\boldsymbol{M}$ and $\boldsymbol{H}$, and does not appear to have been recognized previously (even though a similar effect can be seen in recent results for Fe wires [16]). The microscopic origin of this magnetoresistance has not yet been identified. While it is a function of the wire diameter [15], it does not appear to be associated with weak localization or electron–electron interaction effects [17].

In an 'ideal' system, i.e. a perfectly uniform, homogeneous wire with no crystalline defects, etc., the walls would presumably be able to move freely along the wire. In such a system, magnetization reversal would occur at a vanishingly small field, and there would be no hysteresis. However, real materials do, of course, have hysteresis, as domain walls do not move freely. Indeed, this is indicated by the fact that a field of ~ 150 Oe was required to initiate the reversal process in figure 1. Evidently (and not surprisingly), walls can be trapped at 'pinning' centres along the wire. For reasons which will be described below, we believe that these pinning sites are width fluctuations. Each pinning site forms a potential well which can trap a domain wall. The central question for our experiment concerns the process by which a wall escapes from this well.

An expanded view of the behaviour at fields in the neighborhood of the hysteresis onset is shown in figure 2. These results were obtained by sweeping $\boldsymbol{H}$ up from large negative values, as along the dotted curve in figure 1. It is seen that the resistance of the wire, R, exhibited abrupt jumps. Since we have already observed that ρ depends on the relative orientation of $\boldsymbol{M}$ and $\boldsymbol{H}$, each jump must correspond to an abrupt increase in the amount of the sample with $\boldsymbol{M} \parallel \boldsymbol{H}$; i.e. the abrupt movement of a domain wall as it escaped from a pinning site. In addition, steps were only observed in this, the hysteretic region, which also implies that they were associated with the domain walls and their motion. Currently, we have no way of estimating with certainty the number of walls present in a sample, although we suspect that it is small, probably only one or two. However, even if several walls are present we believe that each jump in $R(H)$ is due to the motion of a *single* wall, since it seems unlikely that walls separated by several μm, and which are travelling in opposite directions, would move in a correlated manner (more evidence in support of this interpretation is given in [14]). The measurements thus enable us to study the kinetics of individual walls [18].

On repeated measurement, the $R(H)$ curves for a particular sample exhibited the *same* sequence of steps; that is, the steps always had the same size and came in the same order. We therefore believe that each step is due to one particular pinning site in the sample. Different samples exhibited a different pattern of steps, as would be expected, since the locations and strengths of the pinning sites should vary from sample to sample.

Repeated measurements with a given sample, as in figure 2, showed that the precise value of the escape field for a particular pinning site varied a small amount from sweep to sweep. According to the theory, this system can be viewed as a 'Brownian' particle (the domain wall), which is trapped in a metastable potential. The depth of the potential well is determined by the nature of the pinning site and also the magnetic field. As the field is increased, the depth of the well (e.g., the barrier height) decreases monotonically, since it must vanish at large fields. Hence, as the field was swept upwards in figure 2, the barrier height decreased continuously until at some point the wall was able to escape from the well. For a Brownian particle this escape should be a stochastic process, which accounts for the variation of the escape field observed with repeated measurements. Other results support

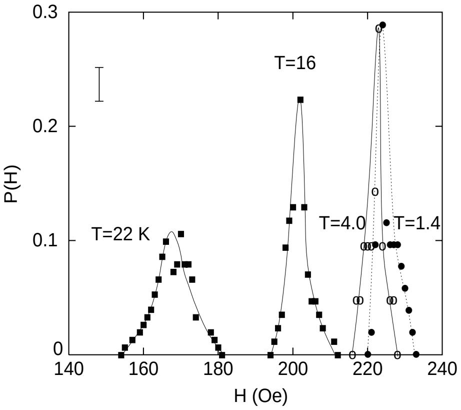

Figure 3. Distribution of escape fields for sample 1 (the sample considered in figure 2) at several temperatures. A typical uncertainty, due largely to the statistical errors, is shown by the bar. The smooth curves are guides to the eye.

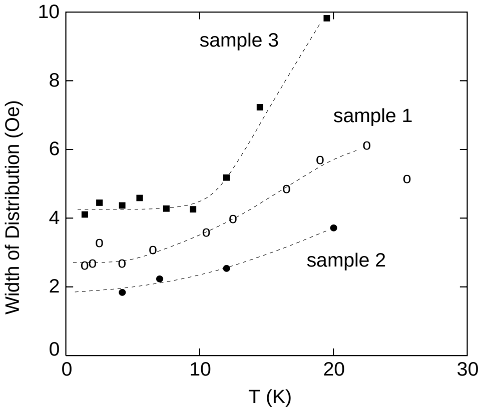

Figure 4. Widths of the escape field distributions for three different samples. Samples 1, 2, and 3 had (average) diameters of 300, 420, and 350 Å, respectively. The dashed lines are guides to the eye.

this picture. For example, the average escape field shifts to larger fields as the sweep speed is increased [14].

In view of the stochastic nature of the escape process, it is clearly desirable to measure the *distribution* of escape fields, $P(H)$, and some results of this kind are shown in figure 3. A single escape field measurement, as in figure 2, required $\sim$ 10 min. This limited us to only 100–200 events per distribution, so the statistical errors were significant. In particular, we do not believe that features such as the apparent 'split peak' in the distribution at 22 K, and the shoulders in the distributions at 4.2 K and 1.4 K are real, since they are comparable in size to the statistical uncertainties. Despite these uncertainties however, certain features are clear [19]. Above $\sim$ 10 K, $P(H)$ broadened and shifted to lower fields as the temperature was increased, while below about 5 K, $P(H)$ was nearly temperature independent. Results for the rms width of the distribution, ΔH, for three different samples, are shown in figure 4. In each case the width increased with T at high temperatures, and appeared to approach a nonzero value as $T \rightarrow 0$. The precise value of ΔH, and also its temperature dependence, varied somewhat from sample to sample, presumably because of differences in the shapes of the pinning barriers, etc.

It is natural to suppose that the effective particle, i.e. domain wall, can be thermally activated over the potential barrier associated with a pinning site. At high temperatures the thermal energy of the particle will be relatively large, enabling it to escape over relatively high barriers. As T is decreased, a wall will (on average) only be able to escape over barriers which are lower. Since the barrier height is smaller at higher fields, this explains why the average escape field becomes smaller as T is increased. In addition, for thermally activated escape the width of $P(H)$ is proportional to T, which accounts for the broadening of the distributions above $\sim$ 10 K. We therefore conclude that at high temperatures the escape takes place by thermal activation over the pinning barrier. However, if thermal activation were the dominant escape mechanism over the entire temperature range, then ΔH should vanish as $T \rightarrow 0$. This is clearly not the case. According to the theory, one should expect the escape to occur via tunnelling through the barrier (MQT) at low temperatures. This mechanism yields a temperature independent ΔH, as observed at low temperatures in figure 4.

Our results are thus in good general accord with the theory, which predicts that domain walls should escape from pinning centres via thermal activation at high temperatures, and MQT at low temperatures. The results for $P(H)$ are central to this conclusion. If, as we have argued, each step in the $R(H)$ curves is due to wall escape from a particular pinning site, it is very hard to see how to explain the behaviour of ΔH without appealing to MQT. However, there are a number of unresolved questions. First, the theory [20] predicts that the crossover from thermal activation to MQT should occur at ~ 0.4 K; i.e. about an order of magnitude lower than we observe [21]. This crossover temperature is determined by the frequency for small oscillations in the potential well, and thus depends on the shape of the well [22]. The theoretical assumptions regarding this shape (which seem quite reasonable) may need to be reexamined. In any event, given the uncertainties at present, we view this level of agreement as quite encouraging. Second, the question of 'dissipation' [1, 5, 6], i.e. the interaction of a wall with its environment, and how this affects the tunnelling rate, remain (experimentally) to be explored. Third, the results for $P(H)$ can be used to obtain the escape rate as a function of H [23, 24]. Results of this kind are essential for a complete determination of the tunnelling parameters, and will be presented elsewhere. Fourth, the nature of the pinning sites is not completely clear, but we believe that they are width fluctuations, for the following reason. We have been able to lithographically introduce abrupt, yet controlled changes in the width. Such samples exhibit much larger resistance jumps than nominally uniform wires. In fact, the three samples considered in figure 4 had these intentionally produced pinning sites [25]. Finally, while we believe that our transport measurements provide strong support for the picture of domain wall motion described above, it would clearly be desirable to confirm this interpretation through other types of measurements. We are optimistic that more direct observations through, e.g., magneto-optic microscopy, are feasible.

In conclusion, we have used transport measurements to study the motion of domain walls in a quasi-one dimensional ferromagnet. The results suggest that the walls escape from pinning sites by thermal activation at high temperatures and MQT at low temperatures. A particularly attractive feature of this experiment is that we seem to be observing the behaviour of an *individual* tunnelling entity. However, further work will be needed to verify that this is, in fact, the case.

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[2] For a review see Clarke J, Cleland A N, Devoret M H , Esteve D, and Martinis J M 1988 *Science* **239** 992
[3] Giordano N 1994 *Physica* B **203** 460
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[6] Stamp P C E, Chudnovsky E M and Barbara B 1992 *Int. J. Mod. Phys.* B **6** 1355
[7] Recent work on magnetic MQP is reviewed in L Gunther and B Barabara (eds.) 1995 *Quantum Tunneling of the Magnetization–QTM '94* (Dordrecht: Kluwer). The evidence for various types of macroscopic quantum phenomena in magnetic systems is not universally accepted, see Garg A 1995 *Phys. Rev. Lett.* **74** 1458 and references therein.

[8] Notable exceptions are the experiments of Lederman M, Schultz S and Ozaki M 1994 *Phys. Rev. Lett.* **73** 1986 and Wernsdorfer W, Hasselbach K, Mailly D, Barbara B, Benoit A, Thomas L and Suran G 1995 *J. Mag. Magn. Mater.* **145** 33 where the magnetization reversal of individual submicron particles was studied.

[9] Prober D E, Feuer M D and Giordano N 1980 *Appl. Phys. Lett.* **37** 94

[10] Typical sample currents were a few μA. Heating due to the sample current or spurious noise is a major concern in experiments of this kind. We were able to check for this by examining the electron–electron contribution to the resistance. This contribution had the expected form and magnitude down to our lowest temperature (1.3 K), indicating that the electrons were indeed cooled to this temperature.

[11] This is the thickness of domain walls in bulk Ni (see, for example, [12]). In our polycrystalline samples, this will probably be a lower bound on the wall thickness [12]. We should hasten to add that the structure of the domain walls in our wires may be more complicated than the usual picture of a Bloch or Néel wall. In addition, while the demagnetizing energy will tend to make M lie along the axis, there may be some deviations ('splay') in direction near the ends or edges of the sample (and near the walls, of course). However, we do not believe that this will make the wall thickness less than the sample diameter. It still seems safe, as a first approximation, to think of domains with M directed along the axis of the wire, separated from each other by some sort of wall or 'transition region'. A detailed theoretical analysis will, of course, have to deal with these complications, but since our sample geometry is well characterized, we feel that such an analysis should be feasible.

[12] Jiles D 1991 *Magnetism and Magnetic Materials* (London: Chapman and Hall)

[13] One might also imagine that walls could nucleate in pairs at sites along the wire. However, experiments comparing the behaviour with opposite field polarities [14] allow us to rule out this possibility at the fields (near the minimum in the $R(H)$ curve) considered in detail here. We hope to search specifically for nucleation effects in future experiments.

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[17] Lee P A and Ramakrishnan T V 1985 *Rev. Mod. Phys.* **57** 287

[18] From measurements of the hysteresis, as in figure 1, it is possible to use the size of an $R(H)$ jump to estimate how far the wall has moved. The jumps in figure 2 correspond to a wall displacement of ~ 1.2 μm.

[19] The results for $P(H)$ were stable with time, and were unaffected by thermal cycling, even to room temperature. The results shown in figure 4 all correspond to the first jump in $R(H)$, but jumps at higher fields exhibited similar behaviour.

[20] This estimate of the crossover temperature was obtained using equation 3.67 in [6].

[21] It is interesting that studies of wall motion in bulk samples, using a rather different approach, also infer larger values of the crossover temperature than expected theoretically; see Barbara B, Wegrowe J E, Sampaio L C, Nozières J P, Uehara M, Novak M, Paulsen C, and Tholence J L 1993 *Physica Scripta* **T49** 268.

[22] According to the theory [6], the small oscillation frequency and the crossover temperature are functions of material parameters (such as M), sample shape, and the assumed form of the pinning potential.

[23] Kurkijärvi J 1971 *Phys. Rev.* B **6** 832; Fulton T A and Dunkleberger L N 1974 *Phys. Rev.* B **9**, 4760; Garg A 1995 to be published

[24] The relation between the escape rate and the escape distribution is discussed in detail in [23] and [2].

[25] These width variations were such as to make the central half of the sample ~ 50 Å narrower than the end regions. The motivation for modifying the wires in this manner arose from the discussion in Gunther L and Barbara B 1994 *Phys. Rev.* B **49** 3926.