# Spin dynamics of $SrCu_2O_3$ and the Heisenberg ladder

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The $S=1/2$ Heisenberg antiferromagnet in the ladder geometry is studied as a model for the spin degrees of freedom of $SrCu_2O_3$. The susceptibility and the spin-echo decay rate are calculated using a quantum Monte Carlo technique, and the spin-lattice relaxation rate is obtained by maximum-entropy analytic continuation of imaginary-time correlation functions. All calculated quantities are in reasonable agreement with experimental results for $SrCu_2O_3$ if the exchange coupling $J\approx 850$ K. However, for the susceptibility fit an anomalously low factor is required.

The Cu-O layers of $SrCu_2O_3$ have an internal structure of parallel double chains (ladders).[1,2] Cu spins within a ladder are exchange coupled with a strength expected to be comparable to that of high-$T_c$ cuprates, whereas the interladder coupling is weak, arising from 90° Cu-O-Cu bonds. The spin degrees of freedom should therefore be well described by the Heisenberg model on a single ladder,[3] defined by the Hamiltonian

$$\hat{H}=J_1\sum_i \sum_{a=1,2} \vec{S}_{a,i}\cdot\vec{S}_{a,i+1}+J_2\sum_i \vec{S}_{1,i}\cdot\vec{S}_{2,i}, \qquad (1)$$

where $\vec{S}_{a,i}$ is a spin-1/2 operator at site $i$ of chain $a$. It is now well established that this system has a gap between the ground state and the lowest excitation for any ratio $J_2/J_1 \neq 0$. For $J_1=J_2=J$, the gap is $\Delta=0.504J$.[4]

Recent experiments on $SrCu_2O_3$ have been carried out by Azuma *et al.*[2] and Ishida *et al.*[5] Their results for the spin susceptibility $\chi$ and the $^{63}$Cu NMR spin-lattice relaxation rate $1/T_1$ show clear evidence of a gap. Accordingly, the spin-echo decay $1/T_{2G}$ rate saturates at low temperatures, indicating a finite correlation length in the ground state.[5] However, comparing the data for $\chi$ and $1/T_1$ with theoretical low-temperature results for the Heisenberg ladder obtained by Troyer *et al.*,[6] there is a significant discrepancy; $\chi$ indicates a gap $\Delta\approx 420$ K, whereas the behavior of $1/T_1$ suggests a gap close to 700 K.[2,5,6] At first sight, one would tend to believe that the gap extracted from $1/T_1$ is the correct one, since the corresponding value of $J\approx 2\Delta$ is then close to the exchange constants typically found in planar cuprates.

We have carried out quantum Monte Carlo (QMC) simulations of the Heisenberg ladder, and obtained results for the quantities discussed above. Here we present comparisons with the experimental results, and discuss a possible reason for the gap-size discrepancy found in earlier work. We argue that the formula used to extract the gap from $1/T_1$ is not applicable in the temperature regime where it was used, and that the gap obtained from $\chi$ is more accurate. The calculated $1/T_{2G}$ is also in close agreement with the experimental result for $J\approx 850$ K, corresponding to the smaller gap.

Troyer *et al.* calculated $\chi$ and $1/T_1$ for the ladder by considering the magnon dispersions obtained in the limit $J_2\gg J_1$.[6] The lowest branch is a single-magnon state which is odd with respect to interchange of the two chains $(k_y=\pi)$. This remains the lowest excitation also when $J_2=J_1$. The smallest gap $(\Delta)$ is at momentum $k_x=\pi$ along the chains. As $k_x\rightarrow 0$, the one-magnon branch crosses into a multi-magnon continuum. At $k_x=0$ the gap is $\approx 2\Delta$, corresponding to a two-magnon excitation. At low temperatures the thermodynamics of the ladder is thus obtained by populating the modes with $k_x\approx\pi$, $k_y=\pi$. The susceptibility then has the form[6]

$$\chi\sim T^{-1/2}\, e^{-\Delta/T}. \qquad (2)$$

For $T$ up to $\approx\Delta$ this form is in good agreement with results from exact diagonalizations of small systems,[7] as well as quantum transfer matrix results.[6] As mentioned above, the agreement with experimental results for $SrCu_2O_3$ is also good, with a $\Delta\approx 420$ K.[2]

The NMR spin-lattice relaxation rate is related to the dynamic structure factor $S(\vec{q},\omega)$ according to[8]

$$1/T_1=\frac{2}{\hbar}\sum_{\vec{q}} |A_{\vec{q}}|^2 S(\vec{q},\omega\rightarrow 0), \qquad (3)$$

where $A_{\vec{q}}$ is the nuclear hyperfine form factor. At very low temperatures, the main contributions to $1/T_1$ come from momentum transfers $q_x\approx 0, q_y=0$, i.e., both the initial and final states are on the one-magnon branch at $k_x\approx\pi$. Taking into account only these processes, Troyer *et al.* obtained the leading low-temperature form[6]

$$1/T_1\sim |A_{q=0}|^2\, e^{-\Delta/T}. \qquad (4)$$

A behavior close to exponential is seen for $SrCu_2O_3$ in the temperature regime $100\ \mathrm{K}\lesssim T\lesssim 300$ K.[2,5] At lower temperatures $1/T_1$ is dominated by impurity effects. The $J\approx 1300$ K extracted from fits of (4) to experimental data is markedly different from the $J\approx 850$ K obtained from the susceptibility.

One could certainly argue that $SrCu_2O_3$ is not a perfect ladder system. Most likely, $J_1$ is not exactly equal to $J_2$.

 

However, the above theoretical forms only depend on the gap, and the disagreement between the gaps from $\chi$ and $1/T_1$ cannot be explained by $J_1 \neq J_2$ alone. Furthermore, the coupling between the ladders is expected to be weak.[3] Thus, before discarding the single ladder as a good approximation of the system, it is important to investigate its behavior in more detail. Whereas the low-temperature form (2) for the susceptibility has been verified to be accurate by comparisons with numerical results,[6,7] the form (4) for the spin-lattice relaxation rate has not been tested numerically. At very low $T$ it is hard to see why (4) should not apply. However, the temperatures for which the fit to the experimental results were made are not very low on the scale set by the gap. It is clear that there will be large contributions to $1/T_1$ from processes with $q_x \approx \pi, q_y = \pi$ between the one-magnon branch and the continuum at $k_x \approx 0$ if the temperature is high enough for states at energies $\gtrsim 2\Delta$ to be populated. These processes are particularly important because the ladder has strong short-range antiferromagnetic correlations. The matrix elements entering the $q_x \approx \pi, q_y = \pi$ processes are therefore much larger than those for $q_x \approx 0, q_y = 0$. Hence, although the $q_x \approx 0, q_y = 0$ contributions are the only ones surviving in the $T \rightarrow 0$ limit, it is quite likely that $1/T_1$ is actually dominated by other processes at the upper range of temperatures considered in the experiments.

We have calculated $1/T_1$ using the maximum entropy (ME) method[9] to analytically continue imaginary-time correlation functions obtained by a QMC technique. The spin-echo decay rate $1/T_{2G}$ is related to the static susceptibility,[10] which can be calculated directly. We have used a recently developed QMC method based on stochastic series expansion,[11] which produces results free from systematical errors associated with Trotter based methods.

The calculations of $1/T_1$ and $1/T_{2G}$ require knowledge of the Cu nuclear hyperfine interactions. For the high-$T_c$ cuprates $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ the hyperfine couplings are well described by the Mila-Rice form,[12] with an anisotropic on-site coupling with components $A_\perp$ and $A_\parallel$, and an isotropic nearest-neighbor transferred coupling $B$. Typical values reported are $B = 41$ kOe$/\mu_B$, $B/A_\perp = 1.2$, and $B/A_\parallel = -0.25$.[12] Knight shift measurements on $SrCu_2O_3$ by Ishida *et al.*[5] indicate that $B$ is much smaller in this compound. Assuming that a single ladder picture is appropriate for the spin system as well as for the hyperfine couplings, and that the $B$ couplings have equal strengths along and across the chains, the Knight shift results[5] give the relations

$$A_\perp + 3B_\perp = 48 \text{ kOe}/\mu_B, \tag{5a}$$

$$A_\parallel + 3B_\parallel = -120 \text{ kOe}/\mu_B, \tag{5b}$$

where we have not excluded an anisotropic $B$. Assuming that the on-site couplings remain close to their standard Mila-Rice values, the transferred couplings in $SrCu_2O_3$ are thus $B_\perp \approx 4$ kOe$/\mu_B$, and $B_\parallel \approx 15$ kOe$/\mu_B$. Considering experimental uncertainties, these estimates are probably consistent with $B_\perp = B_\parallel$. In any case, the magnitude of $B$ is much smaller than the typical two-dimensional cuprate value.

Below we present results for $\chi$, $1/T_1$, and $1/T_{2G}$. For the NMR rates we use the relations (5b) and several values of the ratios $B_\perp/A_\perp$ and $B_\parallel/A_\parallel$. We consider two choices for the exchange $J$, corresponding to approximately the values obtained before from $\chi$ and $1/T_1$; $J = 850$ K and $J = 1200$ K.

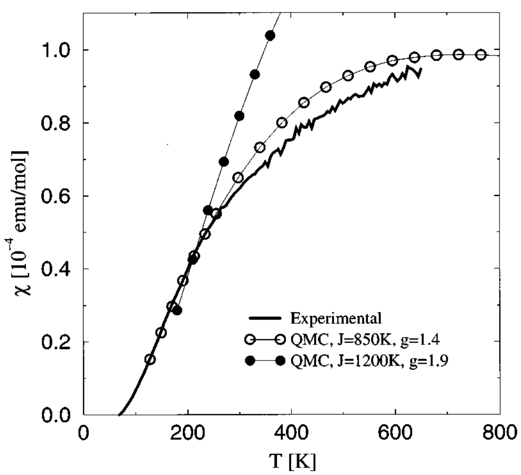


FIG. 1. QMC results for the spin susceptibility of the Heisenberg ladder with $J = 850$ K and $J = 1200$ K compared with the experimental results for $SrCu_2O_3$.

Figure 1 shows our results for the spin susceptibility along with the experimental results by Azuma *et al.*[2] We have used lattices with up to $N = 2 \times 128$ spins, which for the temperatures considered here is enough for finite-size effects to be negligible. The agreement with earlier numerical results[7,6] is good. We adjust the $g$ factor such that the experimental and numerical results agree at $T = 200$ K. For $J = 1200$ K this requires $g \approx 1.9$, but the experimental temperature dependence is not reproduced. For $J = 850$ K the numerical curve matches the experimental data reasonably well over the whole temperature regime, but with an anomalously low $g \approx 1.4$. We note that a small $g$ factor ($g \approx 1.6$) is also needed to match the susceptibility of the linear chain system $Sr_2CuO_3$ to that of the Heisenberg chain.[8]

For extracing $1/T_1$ we have calculated the $r$-space imaginary-time correlation functions corresponding to Eq. (3), and continued these numerically to real frequencies using the ME technique.[9] We have obtained the relevant correlation functions to within relative statistical errors of $10^{-4} - 10^{-3}$ for systems with up to $2 \times 128$ spins. Even with this high accuracy the continued functions have some uncertainties. At high temperatures the procedures can be tested against exact diagonalization results, since the distribution of $\delta$ functions that represent the dynamic structure factor of a small system then is dense enough that a small broadening produces a smooth function, which can be compared with the results obtained with the ME technique. As the temperature is lowered, the number of $\delta$ functions with significant weight decreases rapidly. For the largest systems that can be exactly diagonalized the presence of many gaps then prohibit meaningful comparisons with ME results, since this method cannot resolve structure on that scale. At temperatures where comparisons are meaningful, the ME method produces results in good agreement with exact results for a 16 site Heisenberg chain.[13] Additional evidence that this is a reliable method for obtaining $1/T_1$ stems from work on the two-

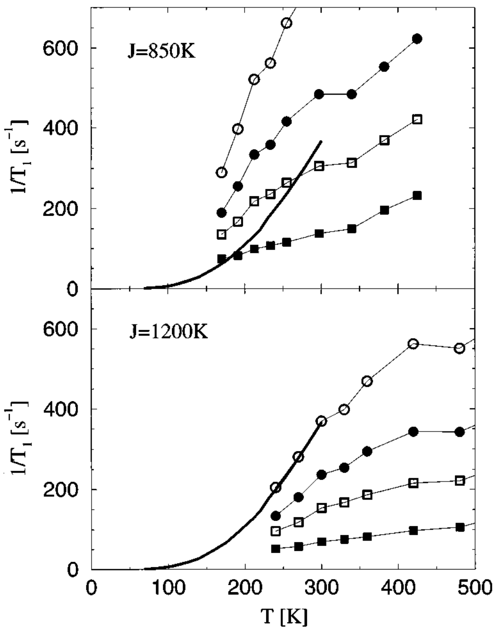


FIG. 2. The spin-lattice relaxation rate calculated using QMC and ME compared with the experimental results by Ishida *et al.* (Ref. 5) (thick solid curves). The upper and lower panels show results for $J=850$ K and $J=1200$ K, respectively. The hyperfine couplings used satisfy (5a). The ratios $B_\perp/A_\perp$ are 0 (open circles), 0.05 (solid circles), 0.10 (open squares), and 0.20 (solid squares).

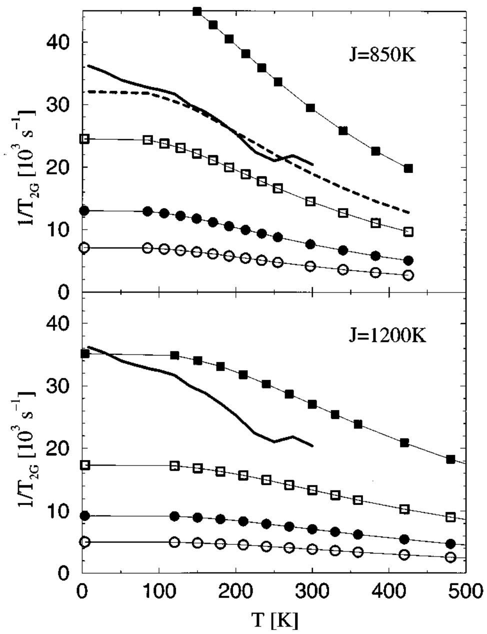


FIG. 3. QMC results for the spin-echo decay rate compared with the experimental results by Ishida *et al.* (Ref. 5) (solid curves). The upper and lower panels show results for $J=850$ K and $J=1200$ K, respectively. The hyperfine couplings used satisfy the relation (5b). The ratios $B_\parallel/A_\parallel$ are 0 (open circles), $-0.05$ (solid circles), $-0.10$ (open squares), and $-0.15$ (solid squares). The dashed curve is the best fit for $J=850$ K, with $B_\parallel/A_\parallel=-0.12$.

dimensional Heisenberg model,[14] where good agreement with experiments on $La_2CuO_4$ was found, as well as results for the one-dimensional Heisenberg model,[13] which exhibit the behavior expected on theoretical grounds.

For the ladder, results obtained using the ME technique become uncertain at temperatures where the gap opens up, and the weight for $\omega\approx 0$ relative to the weight for $\omega>\Delta$ decreases rapidly. We believe that our results are accurate for $T\gtrsim\Delta/2$, and become increasingly inaccurate for lower $T$. Here we present results for $T/J\geqslant 0.2$. The accuracy of the results are probably not higher than tens of percent in the worst cases. Nevertheless, they are useful for establishing the general trends.

Figure 2 shows results for several values of the ratio $B_\perp/A_\perp$, with relation (5a) satisfied. Interestingly, for a strictly local interaction ($B_\perp/A_\perp=0$) and $J=1200$ K there is very good agreement with the experiment. However, in this case (5a) gives $A_\perp\approx 48$ kOe/$\mu_B$, which is much higher than one would expect. It is believed that the on-site couplings should be less sensitive to details of the structure of a particular material than the transferred couplings, and therefore one expects $A_\perp\approx 34$ kOe/$\mu_B$ as in planar cuprates.[12] For $J=850$ K the best overall agreement is obtained with $B_\perp/A_\perp\approx 0.1$, which gives a reasonable value for $A_\perp$ as well. However, the slope of the curve is different from the experimental one. Nevertheless, it is interesting to note that the magnitude of $1/T_1$ agrees with the experimental curve to within a factor of 2 in the regime 150 K $\lesssim T\lesssim$ 300 K, with a $J=850$ K that accounts for the susceptibility as well.

A clear indication that $1/T_1$ in the regime considered here is not dominated by $\vec{q}\approx 0$ processes is that there is a significant decrease in $1/T_1$ with increasing $B_\perp/A_\perp$. With (5a) satisfied, the form factor $A_{q=0}$ remains constant, and hence the low-$T$ form (4) predicts a $1/T_1$ that does not change with $B_\perp/A_\perp$. As we argued above, one can expect processes with $\vec{q}\approx(\pi,\pi)$ to be important at these temperatures, and the decrease in $1/T_1$ with increasing $B_\perp/A_\perp$ is then naturally explained by the decrease in the form factor at $\vec{q}=(\pi,\pi)$.

Only rough estimates of the behavior of the spin-echo decay rate of the Heisenberg ladder have been made.[5] It is dominated by the indirect nuclear spin-spin interactions induced by the coupling to the electronic spin system. Pennington and Slichter derived the form[10]

$$\frac{1}{T_{2G}}=\left[\frac{0.69}{2\hbar^2}\sum_{x\neq 0}J_z^2(0,\vec{x})\right]^{1/2}, \tag{6}$$

where $J_z(\vec{x}_1,\vec{x}_2)$ is the $z$ component of the induced interaction between nuclei at $\vec{x}_1$ and $\vec{x}_2$:

$$J_z(\vec{x}_1,\vec{x}_2)=-\frac{1}{2}\sum_{i,j}A(\vec{x}_1-\vec{r}_i)A(\vec{x}_2-\vec{r}_j)\chi(i-j), \tag{7}$$

and 0.69 is the natural abundance of $^{63}$Cu isotope. The only nonzero hyperfine couplings are $A(0)=A_\parallel$ and $A(1)=B_\parallel$. For a system with a gap, the static susceptibility $\chi(i-j)=\int_0^\beta d\tau\langle S_i^z(\tau)S_j^z(0)\rangle$ decays exponentially with $|\vec{r}_i-\vec{r}_j|$ even at $T=0$. $1/T_{2G}$ calculated for a ladder with $2\times 128$ spins at $T\ll\Delta$ is therefore a good approximation to the $T=0$ result of an infinite system. For this quantity we can thus obtain ground state as well as finite-$T$ results.

Figure 3 shows results obtained using relation (5b) and several ratios $B_\parallel/A_\parallel$. If $A_\parallel$ is to remain close to its value in planar cuprates we need $B_\parallel/A_\parallel \approx -0.1$, which with $J=850$ K indeed gives a quite good agreement with the experimental result. An almost perfect agreement is obtained with $J=850$ K and $B_\parallel/A_\parallel \approx -0.12$. With $J=1200$ K a slightly larger $B_\parallel/A_\parallel$ is needed to produce an approximate agreement with the experiment, but the slope of the numerical curve cannot be reproduced as well as with $J=850$ K. Note that for a strictly local coupling ($B_\parallel=0$) and $J=1200$ K, which gave a good agreement for $1/T_1$ (Fig. 2), $1/T_{2G}$ is almost an order of magnitude too small.

All the above results were obtained with the assumption that the chain coupling $J_1$ is equal to the rung coupling $J_2$. It is important to consider also the more general case of nonequal couplings. Allowing $J_2 \neq J_1$ we find that the best agreement with the susceptibility is obtained with $J_2/J_1 \approx 0.8$, and $J_1 \approx 1100$ K (this requires a $g$ factor $g \approx 1.55$). The results for $1/T_1$ and $1/T_{2G}$ calculated with these parameters show an agreement with the experiments similar to the results in Figs. 2 and 3.

We conclude that the experimentally measured $\chi$ and $1/T_{2G}$ for $SrCu_2O_3$ can be well accounted for by a Heisenberg ladder with $J=850$ K, and the experimentally determined hyperfine couplings. The calculated $1/T_1$ agrees with the experiment to within a factor of 2. The reason for the discrepancies in this quantity could be details of the hyperfine couplings not taken into account here, such as possible differences in the transferred couplings $B$ along a chain and on a rung. $1/T_1$ is a direct measure of the low-frequency spin fluctuation spectral weight, whereas $1/T_{2G}$ is given by a frequency integral. It is therefore likely that $1/T_1$ is more sensitive than $1/T_{2G}$ to slight deviations from the assumed hyperfine relations (5) in the regime where the low-frequency spin fluctuation spectral weight drops rapidly. Hence, we consider the agreement with the experiment to within a factor 2 reasonable. We propose that the reason for the discrepancies reported earlier[2,6] for the gaps extracted from $\chi$ and $1/T_1$ is that contributions to $1/T_1$ arising from processes with momentum transfer $q_x \approx \pi, q_y = \pi$ are important at high temperatures. Since only a narrow range of relatively high temperatures is accessible experimentally, a fit to the low-$T$ form (4) can give misleading results for $\Delta$.

The value of $J$ hence appears to be smaller than the typical values observed in high-$T_c$ cuprates. This is puzzling, since the Cu-O bond structure of the $SrCu_2O_3$ ladders is the same as that of the two-dimensional cuprates.[1,2] One possible explanation for the reduced value is that $J$ represents an effective coupling once interladder effects are taken into account. The weak frustrated ferromagnetic coupling between ladders is expected to enhance the gap[3] and is therefore not a likely mechanism for reducing the effective $J$. On the other hand, a $c$-axis coupling reduces the gap, and may be important in $SrCu_2O_3$. However, preliminary QMC results for the susceptibility of a stack of weakly coupled ladders with $J>1000$ K do not compare as favorably with the experiments as the single ladder result with $J=850$ K shown in Fig. 1. Our single-ladder results are also consistent with a rung coupling slightly smaller than the chain coupling. The best susceptibility fit is obtained with $J_2/J_1 \approx 0.8$ and $J_1 \approx 1100$ K, which leaves $J_2$ at a low $J_2 \approx 900$ K.

The anomalously low $g$ factor needed to reproduce the susceptibility is another puzzling feature. It could in part reflect a reduced local moment associated with the itinerant nature of the Cu-O chains.

*Note added in proof.* In a very recent paper,[15] Johnston suggests that $g=2.1$, and that $J_2/J_1$ has to be as small as $\approx 0.5$ and $J_1$ as large as $\approx 2000$ K. The high-$T$ susceptibility is then well reproduced, but at low temperatures there are significant deviations.

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[1] Z. Hiroi *et al.*, J. Solid State Chem. **95**, 230 (1991); M. Takano *et al.*, Jpn. J. Appl. Phys. **7**, 3 (1992).
[2] M. Azuma *et al.*, Phys. Rev. Lett. **73**, 3463 (1994).
[3] S. Gopalan, T. M. Rice, and M. Sigrist, Phys. Rev. B **49**, 8901 (1994).
[4] E. Dagotto, J. Riera, and D. J. Scalapino, Phys. Rev. B **45**, 5744 (1992); T. Barnes *et al.*, *ibid.* **47**, 3196 (1993); S. R. White, R. M. Noack, and D. J. Scalapino, Phys. Rev. Lett. **73**, 886 (1994).
[5] K. Ishida *et al.*, J. Phys. Soc. Jpn. **63**, 3222 (1994); Phys. Rev. B (to be published).
[6] M. Troyer, H. Tsunetsugu, and D. Würtz, Phys. Rev. B **50**, 13 515 (1994).
[7] T. Barnes and J. Riera, Phys. Rev. B **50**, 6817 (1994).
[8] T. Moriya, Prog. Theor. Phys. **28**, 371 (1962); S. Eggert, Phys. Rev. B (to be published).
[9] J. E. Gubernatis *et al.*, Phys. Rev. B **44**, 6011 (1991); J. E. Gubernatis and M. Jarrell (unpublished).
[10] C. H. Pennington and C. P. Slichter, Phys. Rev. Lett. **66**, 381 (1991).
[11] A. W. Sandvik and J. Kurkijärvi, Phys. Rev. B **43**, 5950 (1991); A. W. Sandvik, J. Phys. A **25**, 3667 (1992).
[12] F. Mila and T. M. Rice, Physica C **157**, 561 (1989); A. J. Millis, H. Monien, and D. Pines, Phys. Rev. B **42**, 167 (1990).
[13] A. W. Sandvik, Phys. Rev. B **52**, R9831 (1995).
[14] A. W. Sandvik and D. J. Scalapino, Phys. Rev. B **51**, 9403 (1995).
[15] D. C. Johnston (unpublished).