decay rate, an upper limit of 5×10^{-22} cm^2 was calculated for the triplet photoionization cross section at 525 nm. This value is 20 times smaller than the one reported by Holzman et al.,[5] but these authors used excitation which extended down to about 472 nm. Perhaps the smaller available kinetic energy, relative to a threshold at 4 eV, with two 525-nm photons results in more loss of carriers by immediate recombination than at the shorter wavelengths.

Crystals were also exposed to stimulated Raman pulses at 467 and 421 nm, which are the third and fourth anti-Stokes lines from liquid nitrogen. At both wavelengths, the fluorescence was essentially a linear function of the intensity, indicating that one-photon excitation from a vibronic level of the ground state was dominant. The photocurrent had an approximate square-law dependence, which is expected for either singlet-photon or singlet-singlet interaction mechanisms for carrier production. From Nakada's values[6] for the absorption coefficient at 421 nm and Kepler's singlet photoionization cross section,[7] the expected photocarrier generation at this wavelength was calculated to be 3×10^6 cm^{-2} for an excitation intensity of 110 W cm^{-2}. A measured value of 3.2×10^6 cm^{-2} was found. Calculations based on the exciton-exciton interaction mechanism and the measured generation coefficient of Silver et al.[8] yielded currents larger by factors from 15 to 75, depending upon the polarization.

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ULTRASONIC PROPAGATION IN $RbMnF_3$ NEAR THE MAGNETIC CRITICAL POINT

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Measurements of the ultrasonic attenuation and velocity near the magnetic critical point in the cubic Heisenberg antiferromagnet $RbMnF_3$ have been performed over a wide range of frequencies. The ultrasonic attenuation coefficient α is divergent at T_C and obeys the law $\alpha\propto\omega^2(T-T_C)^{-0.32\pm0.02}$ for two decades of reduced temperature T/T_C-1.

Considerable theoretical attention has been focused recently on the behavior of the ultrasonic propagation near the magnetic critical point.[1-5] As the transition temperature of a magnetic solid is closely approached, the fluctuations of the magnetization give rise to an increasing cross section for spin-phonon scattering. This manifests itself physically as a growth of the absorption of the ultrasonic waves. Although most theories predict the attenuation coefficient α to be singular at the critical temperature T_C there appears to be no agreement as to the strength of the singularity. Furthermore, it is not yet clear what differences, if any, exist between the critical behavior of α for the ferromagnet and antiferromagnet. The experimental data on these quantities have been sparse. In antiferromagnetic MnF_2 an investigation[6] showed a frequency-dependent anomaly at T_C but no quantitative conclusion could be drawn about the existence of a singularity. Quite recently, measurements of α in the metallic ferromagnet gadolinium[7] have indicated a singularity of the form $\alpha\propto(T-T_C)^{-1.2}$. We present here some results from an experimental investigation of ultrasonic attenuation and velocity near the Néel temperature in the isotropic Heisenberg antiferromagnet $RbMnF_3$. We find that the attenuation is divergent at T_C and is consistent with the power law $\alpha \propto (T - T_C)^{-0.32 \pm 0.02}$.

The perovskite $RbMnF_3$ is unique in that it bears the closet resemblance to the isotropic Heisenberg antiferromagnet of any physical compound.[8] The Mn^{+2} ions are located on a simple cubic lattice and this symmetry is maintained above and below the Néel temperature[9] (distortion $<10^{-6}$). The low magnetic anisotropy field ($H_A \sim 4$ Oe),[10] the occurrence of a remarkably small thermal expansion anomaly near T_C,[9] and the lack of symmetry change on ordering are direct consequences of the weak coupling between the spin system and the lattice. Lattice instability near T_C does not result, the ensuing transition is second order, and thus the behavior of $RbMnF_3$ should be well represented by the antiferromagnetic Heisenberg model.

Measurements of the ultrasonic attenuation near T_C for frequencies from 30 to 150 MHz were performed on a large (~15-mm cube) single crystal of $RbMnF_3$ of high perfection.[11] Attenuation near T_C was measured relative to the attenuation at $T_C + 15$°K at which temperature the critical scattering was negligible. Ultrasonic velocities were measured by the pulse superposition technique[12] from 4.2 to 300°K for ⟨001⟩ and ⟨110⟩ propagation at frequencies from 10 to 70 MHz. The critical behavior of these quantities for ⟨001⟩ longitudinal waves is exhibited in Fig. 1. The temperature at which the attenuation maxima and the velocity minima appear is frequency independent within our sensitivity and defines $T_C = 82.96$°K. All data were taken at thermal equilibrium with temperature stability to better than 5×10^{-4} deg.

Of central interest is the temperature dependence of the attenuation near T_C. We assume that α obeys a law of the form

$$a(T) = A[T/T_C - 1]^{-\zeta}$$

in the critical region. Figure 2 shows a double logarithmic plot of the critical attenuation versus reduced temperature ($T > T_C$) for ⟨001⟩ longitudinal waves at 110 and 150 MHz. This type of plot is most useful in determining the extent of the critical region ($T/T_C - 1 \lesssim 2\times10^{-2}$).

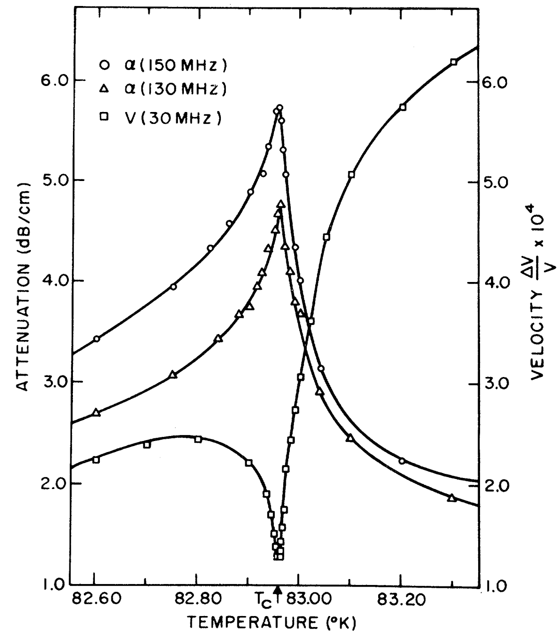

FIG. 1. Critical ultrasonic attenuation and velocity of ⟨001⟩ propagating longitudinal waves in $RbMnF_3$. The attenuation maxima and velocity minima occur at the Néel temperature $T_C = 82.96$°K for all frequencies studied.

The exponent ζ is obtained in practice by choosing the value of ζ which yields the best straight line in a plot of $\alpha^{-1/\zeta}$ vs $T/T_C - 1$. Using the 110-MHz data we find $\zeta = 0.32 \pm 0.02$ for the region $3\times10^{-4} < T/T_C - 1 < 2\times10^{-2}$ with $T_C = 82.96$°K. We find also that the attenuation is quadratic in frequency ($\alpha \propto \omega^2$) over the same temperature range.

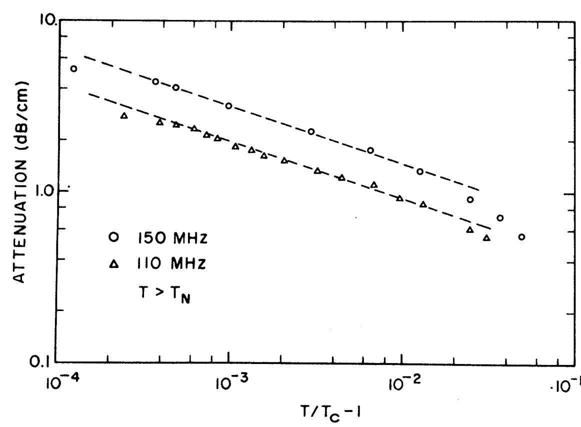

FIG. 2. Double logarithmic plot of attenuation versus reduced temperature ($T/T_C - 1$) for ⟨001⟩ propagating longitudinal waves in $RbMnF_3$. The straight line drawn through the data points corresponds to the critical exponent $\zeta = 0.32$.

By comparing our results for $RbMnF_3$ with those for Gd,[7] we may summarize the experimental situation for the ultrasonic attenuation near the magnetic critical point. (1) The ultrasonic attenuation coefficient is singular at T_C for long wavelengths; (2) the attenuation is proportional to $(T-T_\lambda)^{-\zeta}$ with $\zeta_F > \zeta_{AF}$, where ζ_F and ζ_{AF} are positive critical exponents for ferromagnet and antiferromagnet, respectively; (3) the attenuation is proportional to ω^2 near T_C for both ferromagnet and antiferromagnet.

It does not appear profitable at this point to make too detailed a comparison of the experimental results with existing theory. The attenuation coefficient in the aforementioned theories[1-5] is usually expressed in terms of a four-spin correlation function which is subsequently approximated by two-spin correlation functions. This factorization becomes unreliable near T_C with the net result that the critical fluctuations are overemphasized, leading, in general, to too singular a divergence of α near T_C. It should be noted, however, that the theories of Bennett and Pytte[3] and of Okamoto[4] correctly predict the properties (1)-(3) enumerated above.

Quite recently, a very fruitful approach for predicting the behavior of transport properties near the critical point has been by construction of dynamical scaling laws.[13,14] In particular, it has been proposed that the thermal conductivity of liquid helium should diverge as $(T-T_\lambda)^{-1/3}$ at the lambda point.[13] In particposal, in conjunction with results due to Kawasaki,[15] has prompted the suggestion[16] that in a magnetic solid near T_C the ultrasonic attenuation should be proportional to the thermal conductivity of the spin system, a property whose critical behavior should be obtainable from scaling laws. Although no quantitative theoretical results based on these arguments have been presented as yet, it is to be hoped that the above relationships will be studied in fuller detail. It can be aniticpated also that phenomenological arguments,[17] applied recently to gas-liquid critical phenomena, may find application in magnetic systems and possibly in obtaining the critical behavior of the ultrasonic attenuation.

It is essential to realize that the ultrasonic attenuation is useful as a probe of the spin system only to the extent that the coupling between the spin system and the lattice is well understood. All theories of ultrasonic attenuation have implicitly assumed constant spin-phonon coupling or constant volume at T_C. In a real material in which the lattice parameter may change rapidly at T_C one should expect, in general, significant variations in the effective coupling. In $RbMnF_3$, the volume change is anomalously small so that neglect of coupling changes is reasonable; in other materials this may not be justified.

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