

way as do Ar^+ ions, with slightly higher efficiency. Details of experiments performed with these and other ions will be reported elsewhere.

In conclusion, we have shown that ion neutralization processes at insulator surfaces can cause impurity mobilization in the bombarded films which is a function of the bombarding ion's ionization potential and velocity. In particular, Ar^+ bombardment at 2 keV or less has been shown to cause migration of alkali-metal ions in bombarded SiO_2 films. The consequences for plasma processing, ion implantation, and analysis of insulating films we believe to be far-reaching, in that in all these operations positive ions are impacted on insulating film substrates, and, therefore, impurity migration processes resulting from this bombardment can occur.

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Attenuation of High-Frequency Acoustic Waves on the Low-Temperature Side of the MnF_2 Néel Point

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We present acoustic-attenuation experiments at frequencies up to 1000 MHz in the vicinity of the Néel point of MnF_2 . They show that the attenuation is maximum in the low-temperature phase near the transition, and the temperature at which that maximum appears decreases as the frequency increases. A brief scanning of the theory is given which explains this new effect in terms of relaxation of staggered magnetization.

One of us has recently presented¹ preliminary experiments showing a completely asymmetric behavior of the attenuation of ultrasonic waves around the Néel point of the antiferromagnetic crystal MnF_2 . In the low-temperature phase, the attenuation has a maximum which shifts to lower temperature when the frequency is increased from 100 to 1000 MHz. The temperature shift is as large as 10°K at the highest frequency. In this Letter we would like to summarize new experimental results and present an explanation of this phenomenon in terms of the relaxation of the order parameter, namely, a Landau-Khalatnikov effect. This has very often been invoked to explain anomalous behavior of the acoustic attenuation around a critical temperature²; but the only circumstances where it has been firmly proved is the case of the λ transition of helium.³ We hope to prove here its effectiveness in the case

of the antiferromagnetic transition of MnF_2 .

Theoretical arguments supporting this idea may be given along the following lines. A longitudinally polarized acoustic wave modulates the distance between magnetic ions coupled by exchange forces leading to a Hamiltonian of the form

$$\mathcal{H}_{\text{coupling}} = \sum_{\mathbf{q}, \mathbf{q}'} G(\mathbf{q}\mathbf{q}') \mathcal{E}(-\mathbf{q}) S_{\mathbf{z}}^{(1)}(\mathbf{q}') S_{\mathbf{z}}^{(2)}(\mathbf{q} - \mathbf{q}'),$$

where $G(\mathbf{q}\mathbf{q}')$ is proportional to a derivative of the exchange constant J with respect to the distance between the ions, $\mathcal{E}(\mathbf{q})$ is the strain associated with the acoustic wave, and $S_{\mathbf{z}}^{(1)}$ and $S_{\mathbf{z}}^{(2)}$ are the Fourier components of the magnetization of each sublattice. $\mathcal{H}_{\text{coupling}}$, of course, must include terms involving transverse components of spin operators, but these terms do not enter into the discussion, which is concerned only with the fluctuations of the order parameter. There is a

standard result to note: The attenuation per unit length of the ultrasonic wave⁴ may be expressed as

$$\sigma_1 \approx \omega |G|^2 \chi''(\omega, q),$$

where χ'' is the correlation function of the operator $S_z^{(0)} S_z^{(2)}$ at the frequency ω and wave vector q of the ultrasonic wave.

σ_1 has to be calculated in the two phases; the simplest calculation used a decoupling procedure⁵; in any case it gives an attenuation peaked at T_N , which is more or less symmetric around that temperature and which has been reported in MnF_2 by several investigators.⁶

However, in the low-temperature phase, there is an additional contribution σ_2 to the attenuation due to the fact that

$$\langle S_z^{(0)} \rangle = -\langle S_z^{(2)} \rangle = S(T).$$

$S(T)$ is the order parameter of the antiferromagnet and has a temperature dependence (for $T < T_N$) ϵ^β , where $\epsilon = (T_N - T)/T_N$. σ_2 is simply deduced from σ_1 by a decoupling procedure, which is not an approximation, as

$$\begin{aligned} \sigma_2 &\approx \omega |G|^2 S^2(T) \chi_{N_z}''(\omega, q), & T < T_N, \\ \sigma_2 &\approx 0, & T > T_N, \end{aligned} \quad (1)$$

where $\chi_{N_z}''(\omega, q)$ is now the correlation function

of the staggered magnetization $N_z = S_z^{(0)} - S_z^{(2)}$. χ_{N_z}'' is known from neutron diffraction experiments⁷ and is given by

$$\chi_{N_z}''(q, \omega) = \frac{A_-(T, q)}{q^2 + K_{\parallel}^2(T)} \frac{\omega \Gamma_{\parallel}(T)}{\omega^2 + \Gamma_{\parallel}^2(T)}, \quad (2)$$

where $K_{\parallel}(T) = K_- \epsilon^{\nu'}$ is the inverse of the correlation length, $A_-(T, q)$ is almost constant in the temperature range considered here, and

$$\Gamma_{\parallel}(T) = K_{\parallel}^{3/2}(T) \Omega_-(q/K_{\parallel}) \quad (3)$$

is the characteristic frequency of the magnetization; $\Omega_-(q/K_{\parallel})$ is a scaling function

$$\Omega_-(q/K_{\parallel}) = A + Bq^2/K^2, \quad (4)$$

with A and B experimentally estimated.⁷ Even at the highest frequency used in our experiments and for $\epsilon > 10^{-3}$, $q/K_{\parallel} \ll 1$ so that $\Omega_- \approx A$; in reduced quantities, we may thus finally write

$$\sigma_2(q, \omega) \approx \frac{\epsilon^{2\beta - \nu'/2} \epsilon_0^2}{\epsilon_0^2 + \epsilon^{3\nu'/2}}, \quad (5)$$

where, in former quantities,

$$\epsilon_0 = \omega/\Omega = \omega/AK_-^{3/2}. \quad (6)$$

In the last two articles cited in Ref. 6, σ_1 is clearly resolved at low frequency, and σ_2 begins to appear at intermediate frequency.

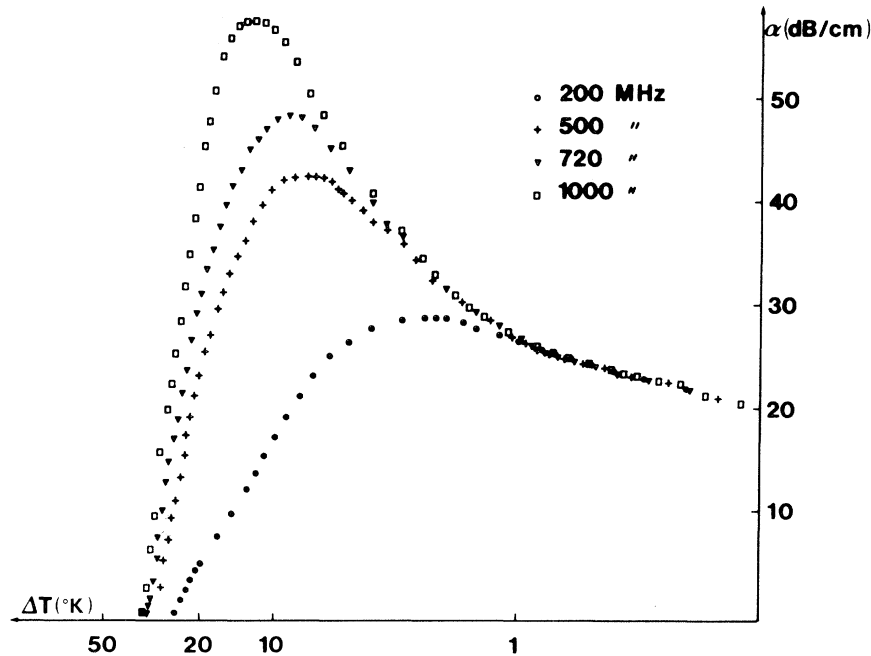


FIG. 1. Experimental curves giving the total attenuation $\sigma_1 + \sigma_2$ in the low-temperature phase at several frequencies. The direction of propagation is (001).

In order to test the formula (5), we propagate longitudinally and transversally polarized acoustic waves along the (100), (110), and (001) axes at frequencies ranging from 100 to 1000 MHz. The MnF_2 crystal⁸ was carefully oriented, polished, and bonded with 4-methyl-1-pentene on a buffer of corundum. Onto the other end of the buffer a ZnO transducer was evaporated and acted as the generator and detector of acoustic waves. The temperature of the sample was carefully controlled and varied by steps.

Typical attenuation curves are given in Fig. 1 for several frequencies. The essential features are the following: The total attenuation $\sigma = \sigma_1 + \sigma_2$ in the low-temperature phase shows a maximum for all the directions of propagation when longitudinal waves are used, but that does not occur for any direction with transversally polarized waves; the amplitude of the attenuation peak depends on the direction of \vec{q} but the shift in temperature does not.

A plot of the temperature T_M for the maximum of the attenuation at different frequencies is given in Fig. 2 and is compared with the theoretical result deduced from (5), $\epsilon_{\max} \simeq \epsilon_0^{2/3\nu'}$; the numerical values⁷ used are $\beta = \frac{1}{3}$, $\nu' = 0.56$, $K = 10^8$ cgs, $A = 3 \times 10^{-2}$ cgs. On the same graph, the second curve gives the values of the attenuation maximum as a function of frequency; the theoretical expression deduced from (5) is $\sigma_{2\max} \simeq \epsilon_0^{(2\beta - \nu'/2)2/3\nu'}$. An analysis of the experimental curve $\sigma_2(T)$ shows that on the low-temperature side of T_M there is some discrepancy with the theoretical expression; it corresponds to the fact that around $T \simeq 50^\circ\text{K}$ the attenuation from the bath of the thermal phonons also decreases. Comparing high- ($T \gg T_N$) and low- ($T \simeq 4^\circ\text{K}$) temperature attenuations, its amplitude may be appreciated; for each frequency the corrected points thus obtained are in closer agreement with the formula (5). The attenuation by thermal phonons also shifts the observed maximum closer to T_N and is probably responsible for the small curvature of the curve $\Delta T(\nu)$ in Fig. 2 at high frequency; for instance, at 1000 MHz the attenuation by the thermal phonons, estimated with the former procedure, is 27 dB/cm; it is to be compared with $\sigma_{2\max}$ which is 57 dB/cm. In that sense a study of a crystal with its Néel point far from the temperature range where the attenuation due to the phonon bath decreases would probably lead to a better agreement.

Finally we have noted that the curves of attenuation are quite independent of any applied magnetic

field up to 30 kG; this rules out the coupling of the acoustic waves with domains which, we know, are suppressed with a few kilogauss,⁹ with nuclear spins¹⁰ whose frequency is limited in MnF_2 at 680 MHz, and with electronic spin waves¹¹ whose frequency is, anyhow, much larger than the frequency of the acoustic waves used in those experiments.

The good agreement obtained between theory and experiment thus seems to show that the Landau-Khalatnikov explanation is very effective. We would like to add some other comments.

(a) It is satisfying to see that this new type of attenuation always occurs with longitudinally polarized waves; because the main exchange interaction between the two sublattices takes place by pairs of Mn^{2+} ions whose axis make an angle with any simple directions of the crystal in which we have propagated acoustic waves, the distance of the ions is always modulated. On the other hand, that does not occur with transverse waves.

(b) The fitting of the curves has been realized without adjusting any parameter. The only liberty used is a numerical value for A entering Γ_{\parallel} which has been chosen within the errors of neutron diffraction results.⁷ As a consequence, in the limit of small wave vector, the relaxation time of the staggered magnetization is expressed as $T_{\parallel}(q \rightarrow 0) = 3 \times 10^{-11} \epsilon^{-2/3\nu'}$.

(c) The behavior of Γ_{\parallel} revealed by those experiments must be emphasized. Scaling laws propose no explicit function for $\Omega_-(q/K_{\parallel})$. Consequently,

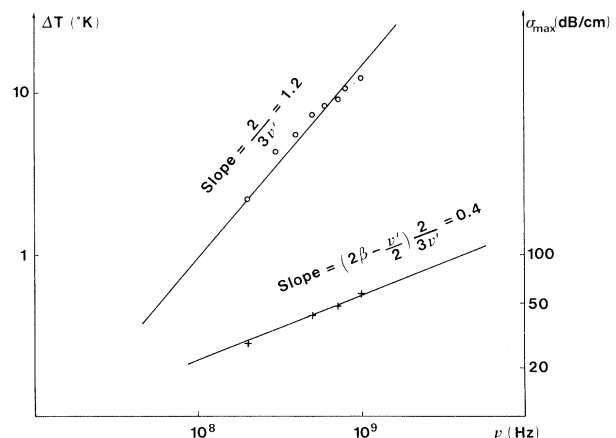


FIG. 2. Circles, experimental points giving the shift in the maximum of amplitude plotted as a function of the wave frequency; a straight line of slope $2/3\nu'$ is drawn for comparison. Crosses, experimental points giving the maximum of the attenuation σ_2 for different frequencies and for longitudinal waves propagating along (001) axis.

Shulhof and co-workers⁷ have analyzed their results successively with

$$\Omega_{\perp} = A + B(q/K_{\parallel})^2$$

and

$$\Omega_{\parallel} = B'(q/K_{\parallel})^2.$$

In fact the values of the q vector spanned by neutron diffraction experiments is always such that $B(q/K_{\parallel})^2 \gg A$ and it is thus difficult to obtain a good numerical value for A . On the contrary, in acoustic experiments, the wave vector q is so small that $B(q/K_{\parallel})^2 \ll A$. Heller¹² has already proposed an explanation for the two contributions to Ω_{\perp} . The first part (A) must be related to the adiabatic longitudinal relaxation. The second part [$B(q/K_{\parallel})^2$] must be related to the isothermal longitudinal relaxation; its characteristic time $1/\Gamma_{\parallel}$ behaves as $1/q^2$ because in the low-temperature phase the magnetization is coupled with the density of energy which itself easily relaxes over small distances (large q) and follows a diffusion law. Following that interpretation, the relaxation time measured in our experiments corresponds to adiabatic fluctuations of the order parameter. It is a common feature in ultrasonic experiments.

In conclusion, the results presented here appear to be complementary to those obtained from neutron diffraction data; they show that the Landau-Khalatnikov effect may also operate in solids.

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E2 Transition Systematics in the *1f-2p* Shell: A Constraint on the Effective Interaction

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We discuss a constraint put by the *E2* transition systematics in the *1f-2p* shell on the effective two-body interaction in the shell. Some simple modifications in the Kuo-Brown effective interaction are made which tend to make it satisfy this constraint.

For a nucleus in the first half of a shell, not suffering from excessive neutron excess, the low-energy states would to a large extent be projectable from a deformed intrinsic state. In this case the magnitudes of the *E2* transitions between such states tend to be proportional to the magnitude of the electric quadrupole moment of the intrinsic state; hence the magnitudes of the experimental values of the quadrupole transition rates can provide some information on the quadrupole

moment of the intrinsic state of the nucleus.

The deformed Hartree-Fock (HF) or Hartree-Fock-Bogoliubov (HFB) state of the nucleus generated by the effective interaction provides a reasonable intrinsic state of the nucleus. The effective interaction should therefore be expected to give rise to intrinsic states for different nuclei such that the quadrupole moments of these have, as a function of the neutron and proton number, a trend similar to that implied by the experimen-