The partial density of vibrational states has been measured for Fe in compressed FeO (wüstite) using nuclear resonant inelastic x-ray scattering. Substantial changes have been observed in the overall shape of the density of states close to the magnetic transition around 20 GPa from the paramagnetic (low pressure) to the antiferromagnetic (high pressure) state. Our data indicate a substantial softening of the aggregate sound velocities far below the transition, starting between 5 and 10 GPa. This is consistent with recent radial x-ray diffraction measurements of the elastic constants in FeO. The results indicate that strong magnetoelastic coupling in FeO is the driving force behind the changes in the phonon spectrum of FeO.
was used, and occurring around 16 GPa \( \Box \) to 17 GPa \( \Box \) under quasihydrostatic conditions.

The calculated partial density of states weighted by the square of the energy \( E^2 \) is plotted versus energy in Fig.3. In Fig.4, the density of states in the Debye approximation would be a straight horizontal line, related to the aggregate sound velocity of the material \( \Box \). The apparent feature of our data is the pronounced softening of the low energy vibrational spectrum. The resolution of the present measurements (2.4 meV) does not allow us to follow the softening to the low frequency region, where most static and ultrasonic measurements are performed. However, this behavior suggests large effects on the static elastic constants at the Néel transition.

The temperature dependence of the elastic constants of Fe\(_{0.93}\)O at ambient pressure was investigated in detail by Sumino et al. \( \Box \). Substantial softening of the shear constant \( C_{44} \) was found close to the Néel transition. Similar anomalies have also been observed in MnO (softening of \( C_{44} \) \( \Box \)). The magnetic structure studies \( \Box \) by neutron scattering have shown that the Mn spins align ferromagnetically within a given (111) plane, and these planes are stacked antiferromagnetically in the <111> direction. Similarly, FeO has the same magnetic structure, with magnetic moments pointing in the direction. Here \( \Delta \) denotes the distortion of trigonally deformed cube netic exchange energy and elastic energy, resulting in a balance is determined by minimizing the sum of mag-

these studies can be summarized as follows. The energy to explain the character of the transition. The results of \[21–23\] studies in MnO invoke exchange-striction effects within the crystal. Experimental \[18–20\] and theoretical \[21,23\] studies in MnO invoke exchange-striction effects to explain the character of the transition. The results of these studies can be summarized as follows. The energy balance is determined by minimizing the sum of magnetic exchange energy and elastic energy, resulting in a rhombohedral distortion

\[
\Delta = z_1 N J_1 \epsilon_1 S^2 / 24 C_{44}
\]

(1)

Here \( \Delta \) denotes the distortion of trigonally deformed cube corner angles \( \frac{1}{4} \pi \pm \Delta \), \( C_{44} \) is the shear constant, \( J_1 \) is the exchange integral for the nearest neighbors, \( z_1 \) is the number of the nearest neighbors, \( \epsilon_1 = -\partial \ln J_1 / \partial r \), and \( S^2 = < S_i \cdot S_j > / < S_i >^2 \) is a correlation function for nearest neighbors with parallel spins (residing within the same <111> plane) and antiparallel spins in neighboring <111> planes. The numerical value of \( \Delta \) is about 0.5 degree at 4 K in MnO. Actually, according to Bartel, \( \Box \) \( \Delta \) is proportional to the sublattice magnetization and is a sensitive measure of the order parameter of the magnetic phase.

However, FeO and CoO have much larger volume anomalies below \( T_N \) and their magnetic properties according to Kanamori \( \Box \) \( \Box \) cannot be described by the theory developed by Lines and Bartel \( \Box \). In a crystal field of cubic symmetry, the orbital degeneracies of Fe\(^{2+} \) and Co\(^{2+} \) are not completely removed, and the residual orbital angular momenta contribute to the energy balance through spin-orbit coupling and the direct effect of orbital magnetic moments, resulting in observable magnetostriction effects. Substantial contribution to the magnetostriction effects in FeO is due to the magnetic anisotropy energy \( \Box \). Kanamori \( \Box \) calculated the equilibrium strain components for FeO resulting from magnetostriction. He used the general formulation derived by Kittel for cubic systems \( \Box \) (all \( C_{ij} \)'s are cubic elastic constants, \( B_1 \) and \( B_2 \) are cubic magnetoelastic constants)

\[
e_{ii} = B_1 ((1/3) - \alpha_i^2) / (C_{11} - C_{12}),
\]

\[
e_{ij} = -B_2 \alpha_i \alpha_j / C_{44}
\]

(2)

where \( \alpha_i \) are direction cosines of the magnetization. Kanamori’s treatment leads to an elongation along [111] diagonal in FeO, which agrees with experimental results \( \Box \). From the experiments on FeO at 95 K \( \Box \), one derives a rhombohedral angle of 59° 32′, or \( e_{xy} = 0.705 \) %. To estimate the rhombohedral distortion resulting from the exchange interaction (Eq.\( \Box \)) in FeO, we need reliable information about the exchange integrals and their dependence on the lattice parameter, which to our knowledge is not available at the moment. The experimental values of distortion \( \Box \) seem to be quite close to the Kanamori’s calculation. Moreover, rhombohedral distortion is enhanced at high pressures \( \Box \), indicating increasing \( B_2 \) in FeO under pressure.

The cubic constant \( B_2 \) is responsible for the magnetoelastic coupling between the phonon and the magnon branches, and our results on the enhanced phonon density of states are directly related to the magnetoelastic coupling in FeO. The expression for phonon dispersion including the magnetoelastic coupling follows from the equations of motion for the magnetic moment \( \Box \) \( \Box \):

\[
(\omega^2 - \omega_m^2)(\omega^2 - \omega_s^2) - \frac{g^2 k^2 (\Omega M_0)}{2 \rho M_0} = 0
\]

(3)

Here \( \omega_s \) is the frequency of sound wave, and \( \omega_m^2 = \Omega g M_0 (\beta + ak^2) \) is the spin wave frequency in the absence of magnetoelastic coupling; \( \rho \) is the density of the material, \( M_0 \) is the magnitude of the magnetic moment of one sublattice, \( \Omega = g M_0 (\beta + 2 \gamma) \). The parameters \( \alpha, \gamma \), and \( \beta \) are related to the exchange interactions and magnetic anisotropy in the material \( \Box \), \( g \) is the gyromagnetic ratio, and \( k \) is a wavevector.

The calculated dispersion relations for FeO at 28 GPa with magnetoelastic coupling included \( \Box \) are shown in Fig.4a (estimated at \( T = 295 \) K), corresponding sound velocities are shown in Fig.4b. Dispersion relations (Fig.4a) change by less than 1 meV, well within the resolution of
neutron inelastic scattering experiment. However, as is evident from Fig.4b, the effect on the sound velocities is more pronounced, being almost 20-30% within the energy transfer range up to 5 meV. This agrees reasonably well with the enhancement of the density of states which we observe in our experiment (Fig.3). Quantitative agreement may be sought along the lines of more elaborated theoretical models, similar to Ref. [4].

At present we do not have enough experimental information and theoretical understanding to better constrain the magnetoelastic coupling at $T/T_N \sim 1$. However, as follows from our observations, the effect is most pronounced close to the Néel transition. Our observations support the Mössbauer measurements, where the magnetic transition was observed starting from 8 GPa under nonhydrostatic conditions. The reason is that uniaxial strain along the body diagonal may induce magnetic moments well below the transition point determined under hydrostatic conditions (17 GPa). This is consistent with the notion that this second order phase transition is smeared by the external field (uniaxial strain), which is proportional to the order parameter of the broken symmetry phase [33]. Further experimental work is required to clarify the effect of exchange-driven magnetoelastic contribution.

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FIG. 1. Inelastic part of the signal in FeO as a function of pressure (elastic peak is subtracted). The region close to zero energy transfer is shown using bold lines. Note enhanced density of states close to $T/T_N \sim 1$. Ratio $T/T_N$ was estimated using $T_N = 198$ K at $P=0.1$ MPa, and data from Ref.[7].

FIG. 2. Dispersion of phonons and magnons in FeO from neutron inelastic scattering experiments. Upper part shows comparison of phonon DOS calculated from neutron measurements (solid line) with our partial DOS for iron at 0.9 GPa (grey circles), 300 K. Also shown are measured and calculated phonon (circles and solid lines) and magnon (diamonds and dashed lines) branches from Ref.[5].

FIG. 3. $g(E)/E^2$ derived from partial DOS in FeO as a function of pressure. For the Debye model, the lower energy part should be a horizontal straight line.

FIG. 4. Model for the magnetoelastic coupling in FeO. a) The interacting transverse phonon and magnon branches. Noninteracting bare frequencies are shown with dashed lines, the dispersion branch for magnons is calculated according to [31], the phonon dispersion was approximated by $E(Q) = 2Qv_0/sin(\pi/2)(Q/Q_0)$ using the sound velocity 3.4 km/sec at $Q=0$. b) Calculated sound velocity including magnetoelastic coupling (solid line) and without magnetoelastic coupling (dash-dotted line).