ler[8] of the field dependence of the spin waves in Tb, the anisotropy-induced wave-vector dependence[6] [Eq. (J8b)] of dE_q^2/dH was not properly accounted for. The A_q and B_q they deduce are therefore not correct. A further analysis is necessary. However, it seems clear that there is only one J_q function in the problem, again making a large two-ion anisotropy superfluous. Jensen refers to an exact numerical analysis for Tb. If it was exact with respect to the spin-wave theory, no further discussion would of course be necessary. Exactness with respect to diagonalizing the single-site part of the Hamiltonian is, in a much simpler and model-independent way, obtained by the MME approach. Jensen finds that the CHF theory can be improved by treating the ground-state correction b [and b' which is proportional to δ' in Eq. (10)] as a fitting parameter. A comparison between Eqs. (2) and (3) and Eqs. (6) and (7) shows that this indeed may approach the CHF result to the MME result. Finally it should be noticed, by comparing (9) and (10), that the CHF theory tends to underestimate the wave-vector dependence of B_q since $\delta' < 1 + \delta$. Jensen's estimate of the effect is thus to be considered as a lower and not upper bound.

The conclusion is that the inclusion of ground-state corrections in the spin-wave theory for strongly anisotropic magnets may be very important and eliminates the need for introducing a giant two-ion anisotropy. It has been demonstrated that the criticism of the MME theory raised by Jensen is invalid. A reliable calculation of the magnitude of the ground-state effects is difficult for the complicated Hamiltonian expected for the heavy-rare-earth metals. An experimental investigation of the ground-state effects in a simple planar magnet would be valuable.

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Temperature Dependence of the Electric Field Gradient in a Quasi-Two-Dimensional Metal: $NbSe_2$†

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The ^{93}Nb nuclear-quadrupole-resonance frequencies have been measured as a function of temperature in 2*H*-$NbSe_2$ between 38 and 376 K. A nearly linear behavior $\nu_Q = a - bT$ is observed between 77 and 376 K in contrast to an approximate $\nu_Q(0)(1-\alpha T^{3/2})$ dependence found in most noncubic metals over the same temperature range. Using a variation of Jena's theoretical treatment of the temperature dependence of ν_Q in noncubic metals, with the consideration that $NbSe_2$ is a quasi-two-dimensional structure, we present an explanation of the nearly linear dependence of ν_Q on T in $NbSe_2$.

Recently, the interest in the mechanism responsible for the temperature dependence of the electric field gradient (EFG), eq, in noncubic metals has been revived by two observations. On the one hand, it was realized that the quadrupole coupling constant, e^2qQ/h, or lowest pure quadrupole frequency, $\nu_Q = 3e^2qQ/2I(2I-1)$, in a number of noncubic metals follows the approximate dependence[1,2]

$$\nu_Q(T) = \nu_Q(0)(1 - \alpha T^{3/2}), \quad (1)$$

with the conduction-electron contribution to the EFG accounting for most of the observed temperature dependence. On the other hand it has been proposed[1] that the temperature dependence arises mainly from the electron-phonon effect on the

crystal potential. This leads to a linear relationship between eq and the Debye-Waller factor. In this note we present accurate measurements of $\nu_Q(^{93}\mathrm{Nb})$ as a function of the temperature in a layered intermetallic compound, $NbSe_2$. ν_Q varies linearly with T over a wide temperature range (77 to 376 K). We suggest that this linear behavior can be viewed as a confirmation of the recently proposed mechanism linking eq to the Debye-Waller factor when one takes into account the effect of the quasi-two-dimensional phonon spectrum appropriate for a layered compound like $NbSe_2$.

The ^{93}Nb nuclear-quadrupole-resonance (NQR) spectra were measured using an induction spectrometer[3] and employing signal-averaging techniques. Frequency scanning through the spectra was accomplished by driving an RF Communication model 808 frequency synthesizer's output frequency with an analog voltage source from the signal-averager's[4] memory channel address. The spectra were accumulated at temperatures between 38 and 376 K employing helium and nitrogen gas blowing techniques.

The $NbSe_2$ samples were prepared in the Ames Laboratory using the iodine-vapor-transport techniques. X-ray analyses of the samples showed a repeat spacing in the crystallographic c direction appropriate for the $2H$ polytype. Evidence for other polytypes or impurity phases has not been observed.

The temperature dependence of the nuclear quadrupole frequency is shown in Fig. 1. Data of ν_Q at various temperatures have been reported previously in the literature.[5-7] However, those measurements were performed in the quadrupole perturbed NMR regime and thus are considerably less accurate than the present data. Among the ν_Q values reported in the literature, we show in Fig. 1 recent measurements which agree, within the experimental errors, with our measurements. Unfortunately, our measurements are extremely difficult to extend to temperatures below 40 K because of the presence of the charge-density-wave transition[8] at $T_c \sim 35$ K which produces a large broadening of the line above T_c and a line splitting below T_c.[9]

Following the conventional notation one can write the EFG as the sum of the lattice ionic contribution and a contribution due to the nonspherical distribution of the background conduction-electron charge density within the unit cell, namely,

$$eq = eq_{\mathrm{latt}}(1-\gamma_\infty) + eq_{\mathrm{el}}, \tag{2}$$

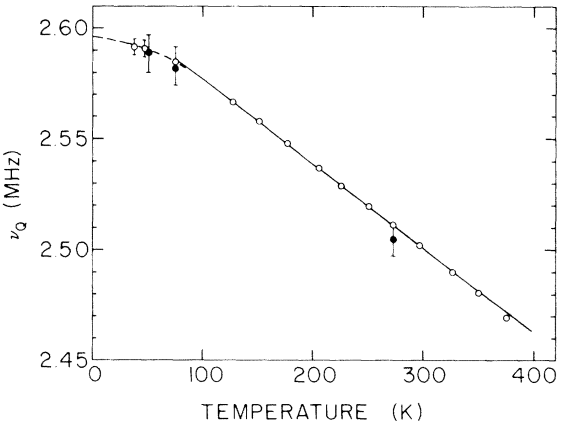

FIG. 1. Lowest pure quadrupole frequency ν_Q of Nb^{93} in $2H$-$NbSe_2$ as a function of temperature. Open circles, present work; closed circles, Ref. (7). The curve drawn through the points helps to visualize the linear behavior. The estimated experimental error in our measurements is of the order of the size of the points with the exception of the two points at low temperatures due to the broadening of the NQR on approaching the charge-density-wave transition temperature.

where γ_∞ is the Sternheimer[10] antishielding factor. Normally, in metals, one finds that the temperature dependence of the ionic contribution, due to lattice expansion, is one or more orders of magnitude too small to account for the experimentally observed temperature dependence. This result is also true for $NbSe_2$ as will be shown below. Therefore, it is believed that the temperature dependence of the quadrupole coupling constant in metals arises mainly from the conduction electron contribution, eq_{el}. This quantity has been expressed by Jena in terms of the temperature as[1]

$$eq_{\mathrm{el}} = eq_{\mathrm{el}}{}^0[1-\beta\varphi(T/\theta_D)], \tag{3}$$

where $eq_{\mathrm{el}}{}^0$ is the value of the EFG at 0 K in the absence of the electron-phonon interaction and $\varphi(T/\theta_D)$ is the function which appears in the Debye-Waller factor:

$$\varphi\left(\frac{T}{\theta_D}\right) = \frac{k_B\theta_D}{3\hbar N 8\pi^3}\iiint \frac{\bar{n}_q+\frac{1}{2}}{\nu_q}\,d^3q. \tag{4}$$

By performing the integral over a Debye sphere, Jena found that the temperature dependence of $\varphi(T/\theta_D)$ and consequently of eq_{el} closely approximates a $T^{3/2}$ dependence for $0<T<2\theta_D$, corresponding to the temperature interval in which the existing measurements in noncubic metals have been made.

For a layered metallic compound like $NbSe_2$

one must take into account the quasi-two-dimensional character of the phonon spectrum. This consideration is important for small wavelengths, namely, for wavelengths shorter than the interlayer spacing. Since the temperature dependence of $\varphi(T/\theta_D)$ is not sensitive to the details of the phonon dispersion relation, we can approximate the effect of the lower dimensionality of the lattice simply by integrating Eq. (4) over a truncated Debye sphere. We indicate by $\nu_D^{\parallel}=k_B\theta^{\parallel}/h$ the maximum frequency of phonons propagating perpendicular to the layer plane and by $\nu_D^{\perp}=k_B\theta_D^{\perp}/h$ the maximum frequency for phonons propagating within the layer (the symbols $\parallel$ and $\perp$ stand for parallel and perpendicular to the c axis). Then we have

$$\varphi\left(\frac{T}{\theta_D^{\perp}}\right)=C_1\left(\frac{T}{\theta_D^{\perp}}\right)^2\int_0^{\theta_D^{\parallel}/T}\left(\frac{1}{e^z-1}+\frac{1}{2}\right)z\,dz+C_2\left(\frac{T}{\theta_D^{\perp}}\right)\int_{\theta_D^{\parallel}/T}^{\theta_D^{\perp}/T}\left(\frac{1}{e^z-1}+\frac{1}{2}\right)dz \tag{5}$$

with $C_1=(\theta_D^{\perp}/\theta_D^{\parallel})[1-\frac{1}{3}(\theta_D^{\parallel}/\theta_D^{\perp})^2]^{-1}$ and $C_2=\frac{1}{2}(\theta_D^{\parallel}/\theta_D^{\perp})C_1$, where the ratio $\theta_D^{\perp}/\theta_D^{\parallel}$ becomes a measure of the two-dimensional (2D) character of the phonon spectrum.

In order to determine the temperature dependence of $\varphi(T/\theta_{\perp})$, we have chosen to plot $\ln[\varphi(\theta/\theta_{\perp})-\varphi(0)]$ versus temperature in Fig. 2 using the notation and the normalization of Jena.[1] The curve for $\theta_{\perp}/\theta_{\parallel}=1$ corresponds to the three-dimensional (3D) case treated by Jena. As it appears in Fig. 2, the curve for $\theta_{\perp}/\theta_{\parallel}=3$ approaches a linear behavior at much lower temperature than for the 3D case. This observation establishes in a semiquantitative way that in a layered metal one should expect a linear temperature behavior of eq_{el} according to Eq. (3) in the same reduced temperature range in which one expects an approximate $T^{3/2}$ behavior for a 3D metal.

Since no theoretical estimates of the quantities eq_{el}^0 and β in Eq. (3) are available, one can only compare the theoretical and experimental temperature dependence by treating the above quantities as adjustable parameters. The ionic contribution to the EFG in $NbSe_2$, calculated by assuming a +5 charge on the Nb atom and −2 on the Se atom,[11] yields a value of $q_{latt}=-0.87$ Å^{-3} at 298 K. The estimate of the lattice contribution to the measured $\nu_Q=\frac{1}{24}(e^2qQ/h)$ depends on a number of poorly known quantities, i.e., Q, γ_∞, and Z, and also on the relative sign of q_{latt} and q_{el}. However, the temperature dependence of q_{latt} and q_{el} is not critically affected by this estimate. We assumed that $|q_{el}|>|q_{latt}(1-\gamma_\infty)|$ and that they are of opposite sign in the light of a recent[12] systematic analysis of the available data in noncubic metals; also we used $Q(^{93}Nb)=-0.2\times10^{-24}$ cm^2 and $\gamma_\infty=-15$. By subtracting the lattice contribution, calculated at different temperatures with the available data on the lattice parameters,[13] and from the measured ν_Q values, we obtain the plot for the reduced $eq_{el}(T)$ shown in Fig. 3. The experimental results are compared with the theoretical predictions of Eq. (3) by choosing for eq_{el}^0

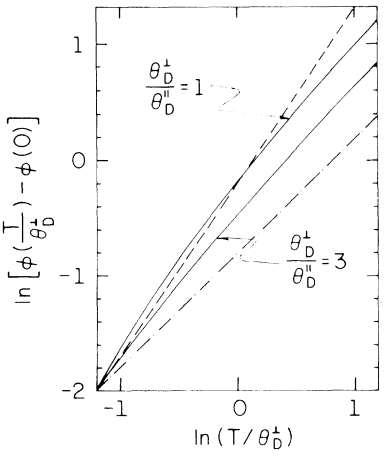

FIG. 2. The solid lines are the plot of $\ln[\varphi(T/\theta_D^{\perp})-\varphi(0)]$ versus $\ln(T/\theta_D^{\perp})$ for two different values of $\theta_D^{\perp}/\theta_D^{\parallel}$. The dashed and dot-dashed curves represent the $T^{3/2}$ and T behaviors, respectively.

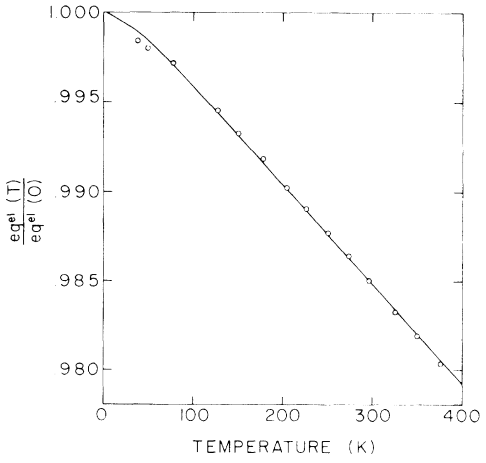

FIG. 3. Plot of the reduced value of the estimated conduction-electron contribution to the EFG in 2H-$NbSe_2$ as a function of temperature. The solid line is the theoretical behavior calculated as explained in the text for $\theta_D^{\perp}=200$ K and $\theta_D^{\perp}/\theta_D^{\parallel}=3$.

and β values which fit the data at $T = 300$ K and at $T \to 0$ K. The theoretical temperature dependence was then estimated by computing $\varphi(T/\theta_D{}^{\perp})$ for $\theta_D{}^{\perp} = 200$ K and $\theta_D{}^{\perp}/\theta_D{}^{\parallel} = 3$ which were chosen in the light of specific-heat[14] and neutron-scattering experiments.[15] The agreement is quite good; however, it should be stressed that the fit with two adjustable parameters is rather insensitive to the exact choice of values for $\theta_D{}^{\perp}$ and $\theta_D{}^{\parallel}$. On the other hand the experimental points in Fig. 3 at low temperatures are quite sensitive to the value chosen for the 0 K extrapolation in Fig. 1.

In conclusion, we have presented new data on the temperature dependence of ν_Q in a quasi-two-dimensional metal which show a different behavior from that of other noncubic metals. We have shown that by assuming the validity of Jena's theoretical explanation for the temperature dependence of eq, the linear behavior can be ascribed to the 2D character of the phonon spectrum in the layered metal. It seems, however, that first-principle calculations of the constants in Eq. (3) are needed, together with accurate measurements in other metals, in order to clearly establish the validity of the mechanism proposed for the temperature dependence of eq.

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