Charge-density-wave state in VS

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The crystallographic phase transition in the compound VS is explained in terms of the formation of a charge-density-wave (CDW) state. In an attempt to neutralize the charge density of the electron gas, the positive ions shift in position and produce a superlattice structure. In VS it is found to be energetically favorable for the superlattice to have a periodicity which is commensurate with the undistorted lattice, as found experimentally. The elementary excitations in the CDW state contain an acoustic-plasmon branch which is mixed with the transverse-optical phonons.

INTRODUCTION

Vanadium and sulfur form the compound VS_{1+x} for a rather wide range of positive and negative values of x. The crystal structure of the compound is the hexagonal NiAs type at high temperatures and the distorted MnP type at low temperatures.[1] The structural phase-transition temperature T_s depends on the value of x.[2] Many other physical properties of the crystal, e.g., the specific heat,[3] magnetic susceptibility,[2] Knight shift,[4] and electrical resistivity,[4] are found to vary drastically at T_s. In a recent publication Liu *et al.* showed by a detailed band calculation that the phase transition may be explained by the formation of a charge-density-wave (CDW) state, and the CDW state is stabilized by the particular Fermi-surface structure of VS in the hexagonal phase.[5]

In this paper we give further discussion of the physical properties of the CDW state as found in VS. The basic theory of CDW state was expounded by Overhauser in a series of articles.[6,7] Halperin and Rice gave an exhaustive review of the many applications of the idea.[8] Therefore, we will not reiterate the theory here, but will concentrate on its application to VS.

In general, the periodicity of the CDW state is determined by the Fermi-surface geometry, in analogy with the spin-density-wave (SDW) state.[9] It is not necessarily commensurate with the lattice structure. We will show by a generalized susceptibility calculation that the Fermi-surface geometry of VS will support a CDW state whose periodicity is very close to twice the lattice periodicity in the basal plane. Then a simple consideration of the strain energy shows that the CDW tends to lock on to the lattice periodicity as observed experimentally. Finally, we show that the elementary excitations in the CDW state include an acoustic-plasmon branch which is mixed with transverse-optical phonons.

ENERGY BANDS, FERMI SURFACE, AND GENERALIZED SUSCEPTIBILITY OF VS

The crystal structure of VS in the high-temperature phase (NiAs type) is shown in Fig. 1. The arrow on each atom shows the direction of displacement of the atom in the low-temperature phase (MnP type). The Brillouin zone of the NiAs-type crystal is shown in Fig. 2. In Ref. 5 it was pointed out that the atomic displacements in the basal plane corresponds to a superlattice with wave vector ΓM. The vanadium atoms in neighboring basal planes also shift in opposite directions, but there is no change in the c axis periodicity of the crystal because each period consists of two V layers. One can visualize the phase relationships of the displacements of V layers as a macroscopic condensation of transverse optical phonons at the zone boundary point and polarized in the a direction. Similarly, the displacements of the S atoms correspond to the condensation of transverse optical phonons at M with polarization vector in the c direction.

The energy bands of VS in the hexagonal NiAs phase is reproduced in Fig. 3. The detail of this calculation will be published elsewhere.[10] The Fermi surface is shown in Fig. 4, from which one can see that the three sheets are largely in the shape of cylinders along ΓA and ML axes. We pointed out in Ref. 5 that the inner cylinder along ΓA and the cylinder along ML are responsible for the stability of the CDW states. The generalized susceptibility function is defined by[11]

$$\chi(\vec{Q}) = \sum_{n,n'} \sum_{\vec{k}} \frac{f_{n\vec{k}}(1 - f_{n',\vec{k}+\vec{Q}})}{\epsilon_{n',\vec{k}+\vec{Q}} - \epsilon_{n,\vec{k}}} , \tag{1}$$

where ϵ_{nk} is the energy of band n and wave vector $\vec{k}$, $f_{n\vec{k}}$ is the Fermi distribution function. The sum on $\vec{k}$ is over the Brillouin zone. Under some simplifying assumptions a peak of this function gives the periodicity of the CDW state. We calculated this function by numerical integration for the two

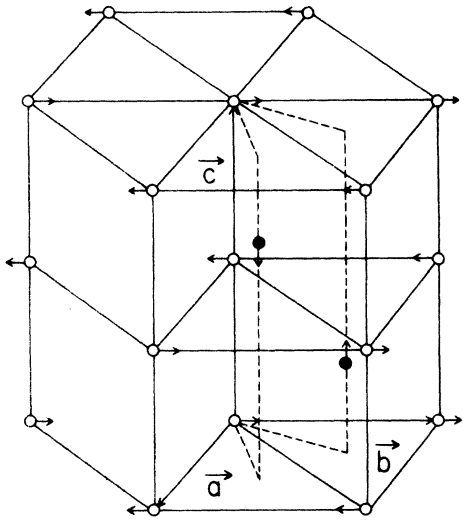

FIG. 1. Lattice structure of VS in the NiAs phase. The open circles are V atoms and the solid circles are S atoms. The arrows on the atoms indicate the direction of lattice displacements in the MnP phase.

bands that give rise to the two important sheets of Fermi surface. The bands were first interpolated to a total of 1 000 000 points in the Brillouin zone, then pairs of band energies $\epsilon_{n\vec{k}}$ and $\epsilon_{n',\vec{k}+\vec{Q}}$ were selected for the sum, where $\epsilon_{n\vec{k}} < E_F$, $\epsilon_{n',\vec{k}+\vec{Q}} > E_F$, and $\vec{Q}$ in the ΓM direction. When the energies differ by less than 0.0001 Ry, the pair was excluded from the sum. The result is shown in Fig. 5. Two curves are presented. The one labeled by $E_F = 0.654$ Ry was calculated using the Fermi level deduced from the integrated density of states.[10] It has a peak at $0.88\,|\frac{1}{2}\vec{\tau}|$, where τ is the reciprocal-lattice vector in the ΓM direction. The curve labeled by $E_F = 0.659$ Ry is for a shifted Fermi level. The reason for this shift is that there are numerous inherent uncertainties in the band calculation so that the energy bands and the Fermi level are uncertain by this order of magnitude. One may notice that this curve has a strong peak at M. The conclusion is that the band structure of VS does tend to stabilize a CDW state, and the characteristic wave vector of the CDW is very close to $\frac{1}{2}\vec{\tau}$.

The observed lattice distortion has the wave vector exactly equal to $\frac{1}{2}\vec{\tau}$. There are two mechanisms which help to lock the CDW to the lattice periodicity. Falicov and Penn[12] showed for the case of spin-density waves (SDW) that in a commensurate structure the two standing waves with wave vectors $\vec{Q}$ and $\vec{\tau} - \vec{Q}$ coalesce, making the commensurate structure stable even though the Fermi surface may give a $\vec{Q}$ slightly away from $\frac{1}{2}\vec{\tau}$. Herring[13] showed that the magnetoelastic interaction between a SDW and the lattice also favors a commensurate structure. These arguments apply equally well to the CDW case.

EFFECTIVE ELECTRON-ELECTRON INTERACTION

In the existing literature on CDW states the electron-electron interaction is treated as a phenomenological potential. We wish to show briefly that the effective interaction consists of a repulsive electrostatic term, an attractive electron-hole correlation term, and an attractive electron-phonon term. The Coulomb interaction operator is

$$V_c = \frac{1}{2}\sum_{\vec{q}} \frac{4\pi}{\epsilon(\vec{q})q^2}\,\rho(\vec{q})\rho(-\vec{q})\,, \tag{2}$$

where $\epsilon(\vec{q})$ is the dielectric function and the charge-density operator $\rho(\vec{q})$ is given by

$$\rho(\vec{q}) = \epsilon \sum_{nn'\vec{k}} \langle n', \vec{k}+\vec{q}\,|\, e^{i\vec{q}\cdot\vec{r}} \,|\, n\vec{k}\rangle \times C^{\dagger}_{n',\vec{k}+\vec{q}}\, C_{n\vec{k}} + \text{H. c.} \tag{3}$$

In Eq. (3), $C_{n\vec{k}}$ is the annihilation operator for an electron in band n and wave vector $\vec{k}$. In the CDW state we have a periodic charge density given by

$$\rho(\vec{Q}) = \epsilon \sum_{k} (\langle 2, \vec{k}+\vec{Q}\,|\, e^{i\vec{Q}\cdot\vec{r}} \,|\, 1\vec{k}\rangle \times \langle C^{\dagger}_{2,\vec{k}+\vec{Q}}\, C_{1\vec{k}}\rangle + \text{c. c.})\,, \tag{4}$$

where 1, 2 are the electron and hole bands that give rise to the nesting Fermi surfaces. The existence of the electron-hole correlation is characteristic of the CDW state.

Strictly speaking, the dielectric function in a multiband metal is a tensor, and the Coulomb screening involves the inverse of the dielectric tensor.[14] We shall continue to write a scalar dielectric function purely for convenience. Furthermore, in $\epsilon(\vec{q})$ we take into consideration all screening effects except that due to the above-mentioned electron-hole correlation. In other words, $\epsilon(\vec{q})$ is

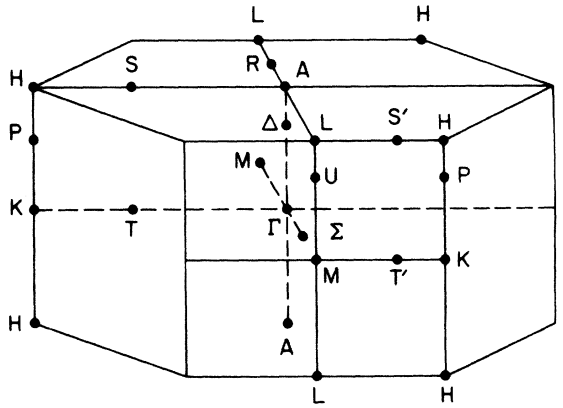

FIG. 2. Brillouin zone of the hexagonal NiAs-type crystal.

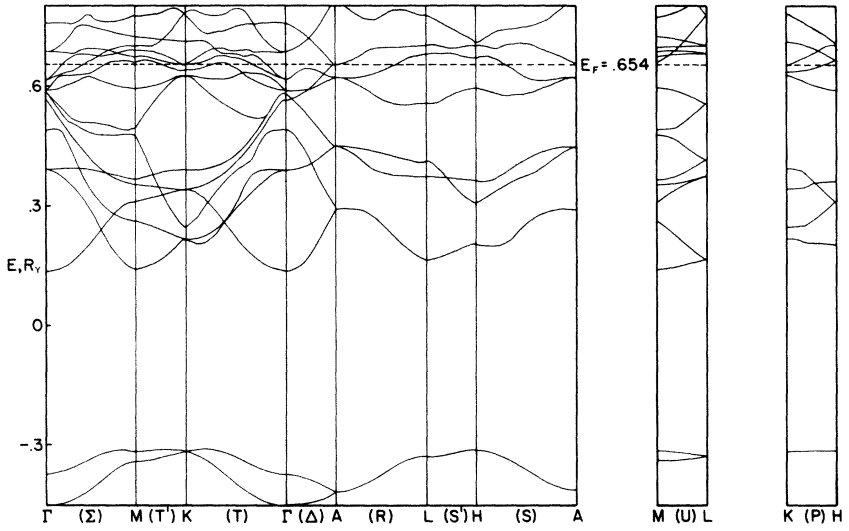

FIG. 3. Energy bands of VS in the NiAs phase.

the total dielectric function of the undistorted phase. The effects of the electron-hole correlation will be discussed later when we study the dynamical properties.

By taking suitable expectation values of the Coulomb interaction operator we find that the CDW state has an excess Coulomb energy

$$V_1 = \frac{4\pi e^2}{\epsilon(\vec{Q})Q^2} \sum_{\vec{k},\vec{k}'} \langle 2, \vec{k}+\vec{Q}| e^{i\vec{Q}\cdot\vec{r}} |1\vec{k}\rangle \times \langle 1\vec{k}'| e^{-i\vec{Q}\cdot\vec{r}} |2, \vec{k}'+\vec{Q}\rangle \langle C^{\dagger}_{2,\vec{k}+Q} C_{1\vec{k}}\rangle \times \langle C^{\dagger}_{1\vec{k}'} C_{2,\vec{k}'+\vec{Q}}\rangle , \quad (5)$$

and a correlation energy

$$V_2 = -\sum_{\vec{k},\vec{k}'} \frac{4\pi e^2}{\epsilon(|\vec{k}-\vec{k}'|)|\vec{k}-\vec{k}'|^2} \times \langle 2, \vec{k}+\vec{Q}| e^{i(\vec{k}-\vec{k}')\cdot\vec{r}} |2, \vec{k}'+\vec{Q}\rangle \times \langle 1\vec{k}'| e^{-i(\vec{k}-\vec{k}')\cdot\vec{r}} |1\vec{k}\rangle \times \langle C^{\dagger}_{2,\vec{k}+\vec{Q}} C_{1\vec{k}}\rangle \langle C^{\dagger}_{1\vec{k}'} C_{2,\vec{k}'+\vec{Q}}\rangle , \quad (6)$$

which arises from Coulomb exchange.

Now we investigate the electron-lattice interaction. For a qualitative description it is convenient to use a rigid-ion model such that the one electron Hamiltonian in a distorted lattice is

$$H = \frac{p^2}{2M} + \sum_{i,\alpha} V_\alpha(\vec{r} - \vec{R}_{i\alpha}) , \quad (7)$$

where i sums over all cells and α sums over all ions in the cell. For a periodically distorted lattice

$$\vec{R}_{i\alpha} = \vec{R}^{(0)}_{i\alpha} + \vec{\epsilon}_\alpha e^{i\vec{Q}\cdot\vec{R}^{(0)}_{i\alpha}} , \quad (8)$$

where $\vec{\epsilon}_\alpha$ is the displacement vector for the αth ion in the unit cell. In the lowest order of the displacement we can write

$$H = H_0 + H_1 ,$$

where H_0 is the one electron Hamiltonian in the undistorted phase and H_1 is the correction due to distortion. In terms of electron operators

$$H_1 = -N \sum_{nn'\vec{k}} \sum_{\alpha} \epsilon_\alpha M_{nn'\alpha}(\vec{k}) C^{\dagger}_{n',\vec{k}+\vec{Q}} C_{n\vec{k}} , \quad (9)$$

where

$$M_{nn'\alpha}(\vec{k}) = \langle n', \vec{k}+\vec{Q}| \hat{\epsilon}_\alpha \cdot \nabla_r V_\alpha(\vec{r} - \vec{R}^{(0)}_\alpha) |n, \vec{k}\rangle \quad (10)$$

and $\hat{\epsilon}_\alpha$ is the unit vector along $\vec{\epsilon}_\alpha$. In the CDW state the expectation value of H_1 is

$$\langle H_1\rangle = -N \sum_{\alpha} \epsilon_\alpha M_{12\alpha}(\vec{k}) \langle C^{\dagger}_{2,\vec{k}+\vec{Q}} C_{1\vec{k}}\rangle . \quad (11)$$

This is the driving term which distorts the lattice. The actual amount of distortion is determined by minimizing the sum of $\langle H_1\rangle$ and the elastic energy.

Since we are working in a region of wave vectors

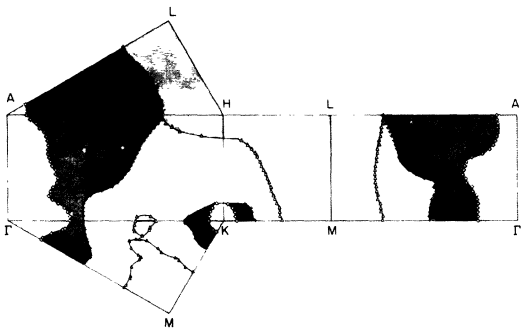

FIG. 4. Fermi surface of VS in the NiAs phase.

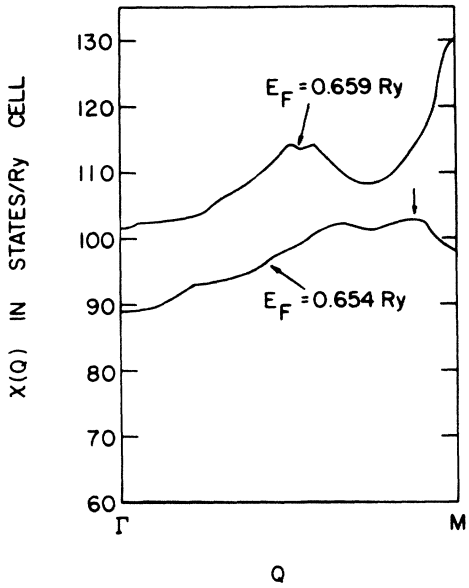

FIG. 5. Generalized susceptibility of VS in the NiAs phase along the ΓM direction.

where the phonon spectrum is relatively flat, we will approximate the lattice by the Einstein model. The expression for the elastic energy is

$$E_L=\frac{1}{2}\sum_{i,\alpha}\lambda_\alpha\left|\vec{R}_{i\alpha}-\vec{R}_{i\alpha}^{(0)}\right|^2 , \tag{12}$$

where the constants λ_α are elastic moduli. For the lattice distortion in Eq. (8), we find

$$E_L=\tfrac{1}{2}N\sum_\alpha\lambda_\alpha\epsilon_\alpha^2 .$$

The minimization of $E_L+\langle H_1\rangle$ yields

$$\epsilon_\alpha=(\lambda_\alpha)^{-1}\sum_{\vec{k}}M_{12\alpha}(\vec{k})\langle C_{2,\vec{k}+\vec{Q}}^{\dagger}C_{1\vec{k}}\rangle \tag{13}$$

and the net elastic energy is

$$E_L+\langle H_1\rangle=-\tfrac{1}{2}N\sum_{\vec{k},\vec{k}'}\sum_\alpha(\lambda_\alpha)^{-1}M_{12\alpha}(\vec{k})M_{12\alpha}^{*}(\vec{k}')\times\langle C_{2,\vec{k}+\vec{Q}}^{\dagger}C_{1\vec{k}}\rangle\langle C_{1\vec{k}'}^{\dagger}C_{2,\vec{k}'+\vec{Q}}\rangle . \tag{14}$$

Adding the results of Eqs. (5), (6), and (14), we obtain the total interaction energy

$$V_1+V_2+E_L+\langle H_1\rangle=-\sum_{\vec{k},\vec{k}'}V_{\vec{k}\vec{k}'}\langle C_{2,\vec{k}+\vec{Q}}^{\dagger}C_{1\vec{k}}\rangle\langle C_{1\vec{k}'}^{\dagger}C_{2,\vec{k}'+\vec{Q}}\rangle , \tag{15}$$

where

$$V_{\vec{k}\vec{k}'}=-\frac{4\pi e^2}{\epsilon(\vec{Q})Q^2}\langle 2,\vec{k}+\vec{Q}|e^{i\vec{Q}\cdot\vec{r}}|1\vec{k}\rangle\langle 1\vec{k}'|e^{-i\vec{Q}\cdot\vec{r}}|2,\vec{k}'+\vec{Q}\rangle+\frac{4\pi e^2}{\epsilon(|\vec{k}-\vec{k}'|)|\vec{k}-\vec{k}'|^2}\times\langle 2,\vec{k}+\vec{Q}|e^{i(\vec{k}-\vec{k}')\cdot\vec{r}}|2,\vec{k}'+\vec{Q}\rangle\langle 1\vec{k}'|e^{-i(\vec{k}-\vec{k}')\cdot\vec{r}}|1\vec{k}\rangle+\tfrac{1}{2}N\sum_\alpha(\lambda_\alpha)^{-1}M_{12\alpha}(\vec{k})M_{12\alpha}^{*}(\vec{k}') . \tag{16}$$

This is the effective electron-electron interaction potential that enters the gap equation.[8] The CDW state is stable at low temperatures if $V_{\vec{k}\vec{k}'}$ is attractive ($V_{\vec{k}\vec{k}'}>0$) for large regions of $\vec{k}$ and $\vec{k}'$ over the bands 1 and 2 near the Fermi level. The first term of Eq. (16) is largely repulsive, and the second and third terms attractive.

PROPERTIES OF DISTORTED PHASE

In the distorted phase an energy gap develops over the nesting portions of the Fermi surface. According to the simple theory the gap depends on the temperature in the same way as described by the BCS theory of superconductivity.[15] The low-temperature gap $2\Delta(0)$ is related to the transition temperature T_s by the simple ratio $2\Delta(0)/kT_s=3.5$.[15] In reality, the gap function is modified by the imperfect Fermi-surface nesting and the phonon and impurity scattering,[16–18] so the situation resembles more closely the antiferromagnetic state of chromium alloys,[18] and we expect $2\Delta(0)/kT_s=4$–5. As an example for $VS_{1.01}$, with $T_s=839$ K,[2] we estimate $2\Delta(0)=0.3$ eV, which should be readily detectable by optical absorption measurements.

The nesting portions of the Fermi surface are destroyed by the energy gap. This causes a sudden decrease of the density of states at the Fermi energy when the temperature drops below T_s. We see evidences of this effect in the 30% drop of vanadium Knight shift,[4] and a similar reduction of the electronic susceptibility.[2] Also, the electrical resistivity increases drastically just bejow T_s, indicating a sudden decrease of the number of carriers. The specific heat has a peak at T_s very similar to that seen at the Néel temperature of chromium.[19]

The gap equation has the form

$$\Delta_{\vec{k}}=\sum_{\vec{k}'}V_{\vec{k}\vec{k}'}\langle C_{2,\vec{k}'+\vec{Q}}^{\dagger}C_{1\vec{k}'}\rangle , \tag{17}$$

where the correlation function $\langle C_{1,\vec{k}'+\vec{Q}}^{\dagger}C_{2\vec{k}'}\rangle$ is related to $\Delta_{\vec{k}'}$. When the phonon and impurity scattering effects are included, Zittartz[17] pointed out that $\Delta_{\vec{k}}$ is the order parameter, not the true gap. In seeking a simple solution one often makes the approximation that

$V_{\vec{k}\vec{k}'} = V$ for $|\epsilon_{n\vec{k}} - E_F|$, $|\epsilon_{m\vec{k}'} - E_F| <$ bandwidth,
$= 0$ otherwise.

Then

$$\Delta = V\sum_{\vec{k}} \langle C^{\dagger}_{2,\vec{k}+\vec{Q}} C_{1\vec{k}} \rangle \ . \tag{18}$$

We may approximate the expression for the lattice distortion [Eq. (13)] by replacing the matrix elements by its average

$$\epsilon_\alpha \cong \frac{\langle M_\alpha \rangle}{\lambda_\alpha} \sum_{\vec{k}} \langle C^{\dagger}_{2,\vec{k}+\vec{Q}} C_{1\vec{k}} \rangle \ .$$

Thus, we find a simple relation between the lattice distortion and the order parameter

$$\epsilon_\alpha = (\langle M_\alpha \rangle / \lambda_\alpha V)\, \Delta \ . \tag{19}$$

This shows that the lattice distortion may be regarded as the order parameter for the phase transition, and it has the temperature dependence like the energy gap of a superconductor or the magnetization curve of an itinerant antiferromagnet. This has recently been confirmed by Franzen's measurements.[2]

COLLECTIVE EXCITATIONS

The collective modes in the CDW state have been found to be charge-density fluctuations (plasmons) with a linear dispersion relation.[20,21] We will add here that since the lattice is involved in the formation of the condensed phase, the plasmons will be mixed with the phonon modes. To study this effect we calculate a dielectric response function and look for self-sustaining modes by examining the analytic properties of this function. It is necessary to simplify the model somewhat so that the mathematics will become tractable without distorting the physical content. The Coulomb part of the interaction (the first two terms of $V_{\vec{k}\vec{k}'}$) will be approximated by a constant V_0. Then the effective Coulomb Hamiltonian has the expression

$$V_c = -V_0 \sum_{\vec{k}\vec{k}'\vec{K}} C^{\dagger}_{1\vec{k}} C^{\dagger}_{2,\vec{k}'+\vec{K}} C_{1\vec{k}'} C_{2,\vec{k}+\vec{K}} \ . \tag{20}$$

The important part of the electron-lattice interaction involves the scattering of an electron from band 1 to band 2 and vice versa, so we write

$$H_1 = N\sum_{\vec{K},\alpha} \epsilon_\alpha(\vec{K}) \langle M_\alpha \rangle \sum_{\vec{k}} (C^{\dagger}_{1\vec{k}} C_{2,\vec{k}+\vec{K}} + \text{H.c.}) \ , \tag{21}$$

where we have extended the results of Eqs. (9) and (13) to include lattice displacements with wave vector $\vec{K} = \vec{Q} + \vec{q}$, and approximated the matrix element by its average. For $\vec{q} \neq 0$ the displacement may be written in terms of phonon operators. For the Einstein model we have

$$\epsilon_\alpha(\vec{k}) = (2Nm_\alpha\Omega_\alpha)^{-1/2} (b^{\dagger}_{\alpha\vec{k}} + b_{\alpha,-\vec{k}}) \ , \tag{22}$$

where $b^{\dagger}_{\alpha\vec{k}}$ creates a phonon with wave vector $\vec{k}$ in the mode α, m_α is the mass of the αth ion, and Ω_α is the mode frequency

$$\Omega_\alpha = (\lambda_\alpha / m_\alpha)^{1/2} \ .$$

In this grossly simplified model of lattice dynamics the mode index is the same as the ion index α. We also adopt a simple band model for the electrons in VS. The model consists of an electron band centered around ΓA and a hole band centered around LM

$$\epsilon_{1\vec{k}} = (1/2m)(k_x^2 + k_y^2) \ ,$$

$$\epsilon_{2\vec{k}} = 2\mu - (1/2m)[k_x^2 + (k_y - Q)^2] \ .$$

The effective masses of the two bands are the same, and the Fermi surfaces are two cylinders of equal sizes.

The calculation of the dielectric response function is very similar to the works in Refs. 20 and 21, and to the calculation of dynamic susceptibility function of an itinerant antiferromagnet.[22,23] We will leave out all details and present only the final answer. We define two dielectric response functions[24]

$$\begin{aligned} \chi_1(\vec{q}, \omega_n) &= \sum_{\vec{k},\vec{k}'} \int_0^\beta \langle T C^{\dagger}_{1\vec{k}}(\tau) C_{2,\vec{k}+\vec{K}}(\tau) C^{\dagger}_{2,\vec{k}'+\vec{K}}(0) C_{1\vec{k}'}(0) \rangle e^{i\omega_n\tau} d\tau \ , \\ \chi_2(\vec{q}, \omega_n) &= \sum_{\vec{k},\vec{k}} \int_0^\beta \langle T C^{\dagger}_{1\vec{k}}(\tau) C_{2,\vec{k}+\vec{K}}(\tau) C^{\dagger}_{1\vec{k}'}(0) C_{2,\vec{k}'+\vec{K}}(0) \rangle e^{i\omega_n\tau} d\tau \ . \end{aligned} \tag{23}$$

The integral equations of these functions are represented by the ladder diagrams in Fig. 6. The solutions may be written

$$\chi_1(\vec{q}, \omega_n) = \frac{1}{2}\left(\frac{\chi_1^{(0)} - \chi_2^{(0)}}{1 - [V_0 + D_0(\vec{K}, \omega_n)](\chi_1^{(0)} + \chi_2^{(0)})} + \frac{\chi_1^{(0)} - \chi_2^{(0)}}{1 - [V_0 + D_0(\vec{K}, \omega_n)](\chi_1^{(0)} - \chi_2^{(0)})} \right) \ . \tag{24}$$

The expression for χ_2 is similar except for a negative sign in front of the second term in the parentheses in Eq. (24). In the above equation the quantities $\chi_1^{(0)}$, $\chi_2^{(0)}$ are the Hartree-Fock approximations to the dielectric susceptibilities, and $D_0(\vec{K}, \omega_n)$ is the phonon propagator given by

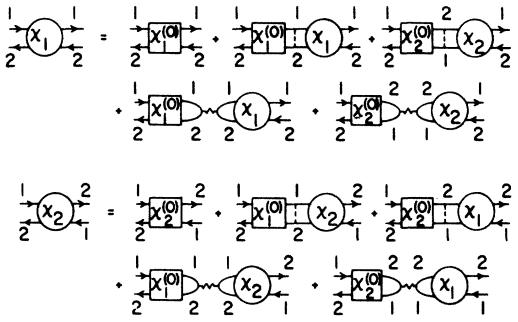

FIG. 6. Diagrams for the renormalized dielectric susceptibility functions in the MnP phase.

$$D_0(\vec{K}, \omega_n) = N \sum_\alpha \frac{\langle M_\alpha \rangle^2}{m_\alpha(\Omega_\alpha^2 + \omega_n^2)} .$$

In the limit $q = 0$, $\omega_n = 0$ one can show that the response functions are singular on account of the gap equation. For small but finite values of q and ω_n, the divergence of the response functions leads to the following dispersion relation of the mixed modes:

$$\omega^2 - c^2 q_\perp^2 + \zeta \sum_\alpha \frac{\langle M_\alpha \rangle^2}{\lambda_\alpha} \frac{\omega^2}{\Omega_\alpha^2 - \omega^2} = 0 , \qquad (25)$$

where $\zeta = 4N\Delta^2/N(0)V^2$, $q_\perp^2 = q_x^2 + q_y^2$, $c = k_F/(2)^{1/2}m$, $k_F = (2mE_F)^{1/2}$, $N(0)$ is the density of states at the Fermi level for each band, and

$$V = V_0 + N \sum_\alpha \frac{\langle M_\alpha \rangle^2}{\lambda_\alpha} .$$

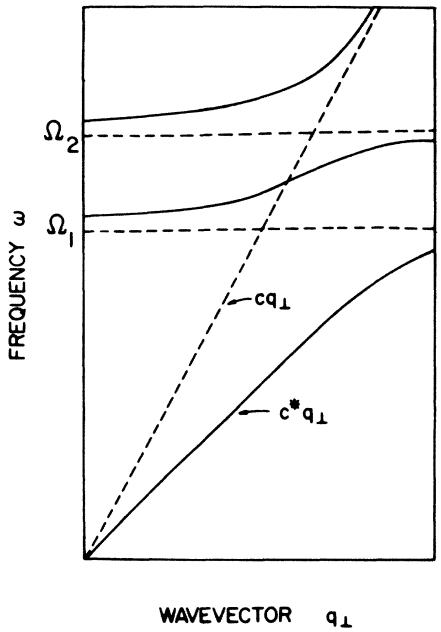

FIG. 7. Schematic diagram of the dispersion curves of the mixed phonon-plasmon modes in the MnP phase.

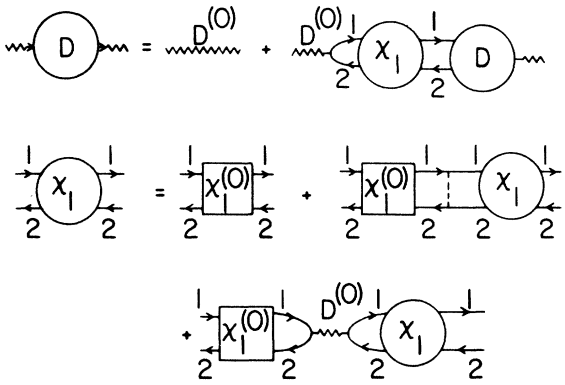

FIG. 8. Diagrams for the renormalized phonon propagator in the NiAs phase.

We have also transformed back to the real frequency domain.

For $cq_\perp \ll \Omega_\alpha$, we get an acoustic branch $\omega^2 = (c^* q_\perp)^2$, where

$$(c^*)^2 = c^2 \Big/ \left(1 + \zeta \sum_\alpha \frac{\langle M_\alpha \rangle^2}{\lambda_\alpha \Omega_\alpha^2}\right) ,$$

and optical branches given by

$$1 + \zeta \sum_\alpha \frac{\langle M_\alpha \rangle^2}{\lambda_\alpha} \frac{1}{\Omega_\alpha^2 - \omega^2} = 0 .$$

The modes are all mixed phonon-plasmon modes. The velocity of the acoustic mode is reduced by the interaction, whereas the optical frequencies are shifted. In Fig. 7 we show schematically the dispersion curves of the mixed modes.

The velocity of the acoustic mode is highly anisotropic because of the Fermi surface geometry. In the real crystal the Fermi surfaces are not perfectly cylindrical, so there will be a small velocity in the c direction.

Above T_s the phonons interact with short-lived charge-density fluctuations, which are analogous to the paramagnons in an itinerant electron antiferromagnet.[25-27] Since phonons are stable for temperatures much higher than T_s, it is more appropriate to study the collective modes above T_s through the phonon propagator. The method of calculation is very similar to that for the dielectric susceptibility functions, so we will leave out the detail and present only the final result. We find by summing the diagrams in Fig. 8 that

$$D(\vec{K}, \omega_n) = D_0(\vec{K}, \omega_n) \times \frac{1 - V_0 \chi_1^{(0)}(\vec{q}, \omega_n)}{1 - [V_0 + D_0(\vec{K}, \omega_n)]\chi_1^{(0)}} .$$

It is easy to verify that $D(\vec{Q}, 0)$ diverges at T_s. At first sight one might take this as an indication that the phonons become soft at the critical tempera-

ture.[28,29] However, a detailed study of the spectral density function of $D(\vec{K}, \omega)$ revealed that just above T_s a peak appears at $\omega = 0$ and $\vec{K} \cong \vec{Q}$. In addition, there are peaks at the optical-phonon frequencies. The zero-frequency peak increases in size as the temperature approaches T_s and $\vec{K}$ approaches $\vec{Q}$, while the optical-phonon frequencies remain constant. Below T_s the zero-frequency peak splits into two at $\pm c^* q_\perp$ and becomes the acoustic-phasmon modes. The optical-phonon frequencies shift upward. The acoustic plasmons and the short-lived charge-density fluctuations appear in the phonon propagator because of the electron-phonon interaction.

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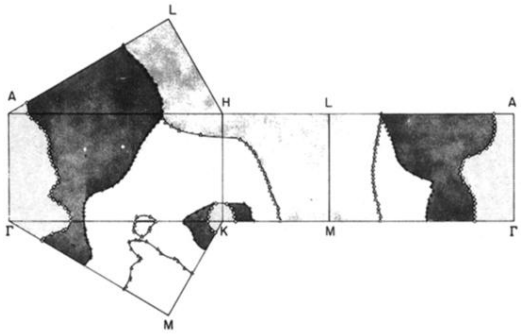

FIG. 4. Fermi surface of VS in the NiAs phase.