

ameters.

The fact that J_{Coul} goes through zero in a number of systems, however, is evidence that \tilde{H}_E is apparently due to phonons in a definite range of impurity concentrations. Numerical calculations have shown that the ratio $|J_{\text{opt}}/J_{\text{Coul}}|$ at $\rho=8$ can vary in a wide range: it is of the order of 0.5 for InSb, AlSb, and PbS, it is equal to 4–6 for many alkali-halide compounds, ZnO, and Cu₂O, and exceeds 10 for CdS, RbI, and CsI. Of course, the contribution of the phonons to J_e increases when account is taken of other mechanisms that couple the electrons with the lattice vibrations (the piezoelectric effect, the optical deformation potential).

Besides the spectroscopic manifestations (the values of the resonance frequencies, the widths of the EPR and APR absorption lines, etc.), interaction via phonons can manifest itself in principles in effects that depend on the magnetic ordering of the impurity. Of interest from this point of view is gallium arsenide, where ferromagnetic ordering of the iron impurity was observed at a concentration 10^{19} cm⁻³ (Ref. 5). At a^* of the order of the lattice constant (~ 5.6 Å), $a \sim 15$ eV, $c_s \sim 4 \times 10^5$ cm/sec, $\rho_0 = 5$ g/cm³, $\epsilon_0 = 11.3$, and $\epsilon_\infty = 10.6$ it turns out that $J_C \approx J_{ac} \approx 118 \times 10^{-3}$ eV. This value of the ex-

change energy accounts fully for the observed ordering temperatures (77–300 K). None the less, this estimate is more readily qualitative, since the deformation-potential method is not valid for impurities with small state radii, and to describe the electron-phonon coupling it is necessary to employ improved procedures.

¹One must not confuse the proposed interaction with that considered by Aminov and Kochelaev.^[1] The latter is of relativistic character and is therefore smaller by several orders of magnitude.

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On the renormalization of the velocity of sound in phase transitions involving a change in valency

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The effect of the hybridization of conduction electrons with a localized f -electron energy level, ϵ_f , lying near the Fermi surface on the longitudinal sound velocity, C , is considered. It is shown that the hybridization enhances the screening of the ion-ion interaction as ϵ_f approaches the Fermi surface. This explains the experimentally-observed minimum in the pressure dependence of the velocity of sound. The strong intrasite correlation of the f electrons leads to an asymmetry in the minimum of $C(\epsilon_f)$ as the ϵ_f level passes through the Fermi surface. The results are compared with the results of the measurement of the velocity of sound in Ce as it undergoes the γ - α transition.

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1. There occurs in a number of rare-earth metals and their compounds (e.g., in Ce and SmS) as the pressure and temperature are varied an electron phase transition connected with the existence of a narrow localized- f -electron band near the Fermi level.^[1] A theoretical model for such a transformation has been worked out in a number of papers^[2–6] on the basis of the transition of f electrons into the conduction band as the pressure is increased.^[2, 3] In this case the presence of strong repulsion between the f electrons at the same time lattice site turns out to be important for the existence of the phase transition.

Later, it was shown^[7, 8] that allowance for the hybridization of the spd - f electrons enables us to account for the intermediate-valence state. It is known that an in-

crease in the compressibility occurs in the vicinity of a phase transition involving a change in valency.^[1] In particular, it has been observed that the longitudinal velocity of sound $C(P)$ in Ce has a minimum in the region of the γ - α transition, which occurs in Ce at a pressure ~ 7 kbar.^[9] The γ - α transition in Ce is iso-morphous, the number of f electrons per site changing in the transition from $n_f = 0.94$ to $n_f = 0.33$.^[10, 11]

In the present paper, using Ce as an example, we show that allowance for the hybridization of the f electrons with the conduction electrons enables us to account for the presence of the $C(P)$ minimum associated with transitions involving a change in valency. This effect has been considered by Kocharyan and Khomskii,^[12] but their approach is purely phenomenological

in character. In contrast to these authors, we develop in the present paper a microscopic theory of this phenomenon with a consistent allowance for the strong correlation of the f electrons localized at the same site. Allowance for this correlation allows the explanation of the experimentally observed asymmetry in the $C(P)$ minimum. Physically, the $C(P)$ minimum turns out to be connected with an increase in the screening of the ion-ion interaction as the f -electron level approaches the Fermi surface. In this case the hybridization transitions of the f electrons into the conduction band increase the density of states near the chemical potential.

Below we first describe the model and the main approximations in which the standard expression for the longitudinal velocity of sound^[3,3] can be used in this model. The main part of the work consists in the computation of the polarization operator. In conclusion, we compare the theory with experiment.

2. Let us consider a system consisting of a lattice of collectivized and localized electrons, taking into consideration the Coulomb interaction between them and the hybridization. The localization of the f electrons leads to their strong repulsion at a site; to describe this repulsion we have to take the f -electron operators in the site representation. The Hamiltonian of the system has the form

$$H = H_{ii} + H_{ie} + H_s + H_{if} + H_{if} \quad (1)$$

$$H_{ii} = \frac{e^2}{2} \sum_{i \neq j} \frac{(Z - n_i)^2}{|R_i - R_j|}$$

$$H_f = \varepsilon_f \sum_{m, \gamma} n_{m\gamma} + \frac{1}{2} U \sum_{m, \gamma, \gamma'} n_{m\gamma} n_{m\gamma'} + \frac{1}{2} \sum_{\substack{n \neq m \\ \gamma, \gamma'}} V(n-m) \Delta n_{m\gamma} \Delta n_{m\gamma'}$$

$$H_s = \sum_{k, \gamma} \varepsilon_k a_{k\gamma}^+ a_{k\gamma} + \frac{1}{2N} \sum_{\substack{k, k', q \neq 0 \\ \gamma, \gamma'}} \frac{4\pi e^2}{q^2 \Omega_0} a_{k+q\gamma}^+ a_{k\gamma} a_{k'\gamma}^+ a_{k'\gamma}$$

$$H_{if} = N^{-1/2} g \sum_{m, A, \gamma} (e^{-ikR_m} a_{k\gamma}^+ + b_{m\gamma} + \text{c.}) + N^{-1/2} \sum_{\substack{q \neq 0 \\ k, \gamma, \gamma'}} v_q e^{iqR_m} \Delta n_{m\gamma} \Delta n_{m\gamma'}^+ a_{k+q\gamma, \gamma'}$$

$$H_{ie} = \frac{1}{N} \sum_{m, A, q \neq 0} v_q e^{iqR_m} a_{k+q\gamma}^+ a_{k\gamma} + \frac{1}{N} \sum_{\substack{q \neq 0 \\ \gamma, \gamma'}} v_q e^{iq(R_n - R_m)} \Delta n_{m\gamma} \Delta n_{m\gamma'}$$

Here we have separated out the mean values of the electron-density operators $\Delta n_{m\gamma} = \hat{n}_{m\gamma} - \langle \hat{n}_{m\gamma} \rangle$. The Hamiltonian H_{ii} describes the interaction of the ions; H_f corresponds to the $(2J+1)$ -fold degenerate single-electron f -electron level, which will be assumed to lie in the middle of a wide conduction band $J=5/2$ in the case of Ce); γ is the total-angular-momentum component; m is the number of the site; U is the intratomic repulsion parameter—the largest energy quantity of the problem (in fact $U \sim 10$ eV). Owing to the separation of the averages of the electron-density operators, the level is assumed, in accordance with the Falicov model,^[4] to be already renormalized by the mean value, v , of the s - f Coulomb interaction $\varepsilon_f = \varepsilon_f^{(0)} + v(n_s - n_f)$. H_{sf} corresponds to the Coulomb repulsion and the s - f hybridization with parameter g . Here, just as in Ref. 7, we assume that the s and f states with the same γ hybridize; v_{is} and v_{if} are the interactions of the electrons with the lattice,

where, for simplicity, the pseudopotentials for the s and f electrons are assumed to be the same:

$$v_{is} = v_{if} = -4\pi e^2 (Z - n_i) / q^2 \Omega_0 + b / \Omega_0$$

The density of states of each of the $2J+1$ conduction bands is assumed to be a constant and equal to ρ .

In the case when the electron-electron interaction does not depend on the position of the lattice sites, the second-order term in the expansion of the Ω potential in powers of the electron-ion interaction,

$$\Omega = \Omega^{(0)} - \langle H_{ie} \rangle + \frac{1}{2} \int_0^{\beta} \langle T H_{ie}(t) H_{ie}(0) \rangle dt$$

gives the well-known expression for the longitudinal velocity of sound^[3,3]:

$$C^2 = \frac{1}{M} \left(- \frac{(Z - n_i)^2}{\Omega_0 \Pi(0, 0)} + \frac{2b(Z - n_i)}{\Omega_0} \right);$$

$$\Pi(0, q) = \langle T (\rho_{q'}^+ + \rho_{q'}) (t), (\rho_{-q'}^+ + \rho_{-q'}) (0) \rangle |_{\omega=0} \quad (2)$$

$$\rho_{q'}^+ = \sum_{\gamma, k} a_{k+q\gamma}^+ a_{k\gamma}, \quad \rho_{q'}^- = \sum_{\gamma, m} \Delta n_{m\gamma} e^{-iqm}$$

Here we have neglected the Umklapp processes.

For the Hamiltonian, (1), under consideration the explicit electron-electron Coulomb interaction function leads to a situation in which the zeroth, $\Omega^{(0)}$, and linear $\Omega^{(1)}$, terms in the power series expansion of the Ω potential in H_{ie} depend on the positions of the lattice sites; consequently, these terms should make an additional contribution to the dynamical matrix. It can, however, be verified that each differentiation of H_f and H_{fs} in $\Omega^{(0)}$ and $\Omega^{(1)}$ with respect to R_m leads in the expression for the dynamical matrix to a superfluous summation over the momentum q (in relation to the analogous differentiation in $\Omega^{(2)}$). Each such summation gives the power of the parameter $\xi^2 = \pi e^2 \rho(\varepsilon_F) / p_F^2$, where $\rho(\varepsilon_F)$ is the density of states at the Fermi surface and p_F is the Fermi momentum.

We shall assume that the parameter ξ^2 is small, and use for C^2 the expression (2). Such an approximation is, generally speaking, valid only in the limit of high electron densities, and should first break down when the level gets sufficiently close to the Fermi surface. But, apparently, a first-order transition with a change in valency occurs when ε_f is still sufficiently far from ε_F . In the case of Ce this is indicated by both a relatively small change in C (by 10–15%), which occurs in the vicinity of the transition, and the closeness of the valency to an integral value (3.06 for the γ phase and 3.67 for the α phase). In the opposite case the expression (2) can be used only in a region far from the transition point.

Finally, let us note that, because of the smallness of the hybridization parameter g as compared to v_{ie} (its characteristic value is $g \sim 0.3$ eV), we can neglect the contribution to the dynamical matrix from the differentiation of the hybridization with respect to the coordinates of the lattice sites.

Within the framework of the above-adopted approximation (i.e., the approximation that $\xi^2 \ll 1$) the determination of C reduces to the computation of the polarization

operator $\Pi(0, 0)$. In the absence of hybridization, $\Pi(0, 0)$ is simply the density of states of the s electrons at the Fermi surface. Physically, it is clear that it is precisely the electron transitions into the conduction band, which occur on account of the hybridization, that should have a significant effect on $\Pi(0, 0)$ and on the total density of states at the Fermi surface. Therefore, in computing $\Pi(0, 0)$ we shall take only the hybridization and the single-site Coulomb interaction U into consideration, and neglect the remaining Coulomb interaction.

3. To determine $\Pi(0, 0)$ it is convenient to use the Ward identity

$$\Pi(0, 0) = - \left. \frac{\partial n}{\partial \mu} \right|_{T, V}, \quad (3)$$

where μ is the chemical potential and n is the total number of particles.

Let us take the correlation interaction U into account at the level of the zeroth Hamiltonian by going over to the atomic-configuration-changing Hubbard operators^[14]:

$$X_m^{00} = \prod_{\gamma} (1 - n_{m\gamma}), \quad X_m^{10} = b_{m\gamma}^{\dagger} \prod_{\gamma \neq \gamma'} (1 - n_{m\gamma'}), \quad X_m^{11} = n_{m\gamma} \prod_{\gamma \neq \gamma'} (1 - n_{m\gamma'}), \quad (4)$$

$$X_m^{\lambda_1 \lambda_2} |\lambda_3\rangle = \delta_{\lambda_1 \lambda_2} |\lambda_3\rangle, \quad X_m^{\lambda_1 \lambda_2} X_m^{\lambda_3 \lambda_4} = X_m^{\lambda_1 \lambda_4} \delta_{\lambda_2 \lambda_3},$$

where $\lambda = |0\rangle, |\gamma\rangle$ and the states $|0\rangle$ and $|\gamma\rangle$ correspond respectively to an empty f shell and a shell with one f electron having a total-angular-momentum component γ . Here we consider the situation in which the single-electron level ε_f with the single f electron lies near the Fermi surface and, consequently, because of the large U value, the single-electron level $\varepsilon_f + U$, which corresponds to the transitions $|\gamma\rangle \rightarrow |\gamma\gamma\rangle$, lies far from ε_F , so that such transitions are not realized.

In the operators (4) the Hamiltonian determining n_s and n_f assumes the form

$$H = H_0 + H_f; \quad (5)$$

$$H_0 = \varepsilon_f \sum_{m, \gamma} X_m^{11} + \sum_{k, \gamma} \varepsilon_k a_{k\gamma}^{\dagger} a_{k\gamma}, \quad H_f = \frac{g}{N^{1/2}} \sum_{m, k, \gamma} (e^{ikm} X_m^{10} a_{k\gamma} + \text{H.c.}).$$

To compute n_f , we apply to the Hubbard operators a technique similar to the one described in Ref. 15, the peculiar form of the technique here being connected with the absence of Fermi commutation relations for the X operators. We shall seek n_f and n_s with allowance for all the terms of first order in the small parameter $\rho g^2 / |\varepsilon_f|$ (the characteristic values $\rho^{-1} \sim 8$ eV, $g \sim 0.3$ eV, and $|\varepsilon_f| \sim g$ give $\rho g^2 / |\varepsilon_f| \sim 0.04$). Below it will become apparent that $\rho g^2 / |\varepsilon_f|$ determines the deviation of n_f from an integral value, so that this approximation corresponds to the case of low f -electron or f -hole density.

Let us represent

$$n_f = \sum_{\gamma} \langle X_m^{11} \rangle$$

in the form of an expansion in powers H_f . For $\langle X^{\gamma\gamma} \rangle$ there arises a series similar to the series for the average spin in the Heisenberg model, a series which is considered in Ref. 15:

$$\langle X^{\gamma\gamma} \rangle = \langle X^{\gamma\gamma} \rangle_0 + \frac{1}{2!} \int_0^{\beta} \int_0^{\beta} \langle T X^{\gamma\gamma}(t) H_f(t_1) H_f(t_2) \rangle_c dt_1 dt_2 + \dots \quad (6)$$

The index c denotes cumulant averaging.

The computation of the X -operator T products is carried out, using the method of Ref. 16, developed for spin operators (see also Ref. 15). The diagrammatic representation of the terms of the series (6) that make contributions of zeroth and first order in ρ has the form

Here the closed and open circles correspond to the operators $X_m^{\gamma\gamma}$ and $K_m^{\gamma} = X_{00}^{\gamma} + X_m^{\gamma\gamma}$ and to an oval corresponds their cumulant averaging; for example,

$$\langle X_m^{\gamma\gamma} K_m^{\gamma} \rangle_c = \langle X_m^{\gamma\gamma} K_m^{\gamma} \rangle_0 - \langle X_m^{\gamma\gamma} \rangle_0 \langle K_m^{\gamma} \rangle_0 = P_0^{\gamma} \delta_{\gamma\gamma} - P_0^{\gamma} K_0^{\gamma};$$

$$P_0^{\gamma} = \langle X^{\gamma\gamma} \rangle_0 = \frac{e^{-\beta\varepsilon_f}}{1 + (2J+1)e^{-\beta\varepsilon_f}}, \quad K_0^{\gamma} = \frac{1}{1 + (2J+1)e^{-\beta\varepsilon_f}}.$$

A continuous line in (7) corresponds to $(i\omega_n - \varepsilon_f)^{-1}$; a dashed line, to the complete Green function $G_s(k, \omega_n)$ for the s electrons:

$$G_s(k, \omega_n) = (i\omega_n - \varepsilon_s - g^2 \Sigma(\omega_n, k))^{-1}.$$

It is shown in Ref. 17 that, correct to second order in $\rho g^2 / |\varepsilon_f|$, we can assume that $\Sigma(\omega_n, k)$ does not depend on momentum. This implies independent scattering of an s electron on the various lattice sites. Thus, in the $\rho = \text{const}$ approximation, the sum over the momenta that enters into the expressions for the diagrams b and c from (7) yields

$$\sum_k G_s(k, \omega_n) = -i\pi \rho \text{sign } \omega_n. \quad (8)$$

The analytic expressions for the diagrams b and c from (7) respectively have the forms

$$-(P_0^{\gamma} - (2J+1)P_0^{\gamma} K_0^{\gamma}) \sum_{k, \omega_n} \frac{G_s(k, \omega_n)}{i\omega_n - \varepsilon_f}, \quad T K_0^{\gamma} \sum_{k, \omega_n} \frac{G_s(k, \omega_n)}{(i\omega_n - \varepsilon_f)^2}.$$

The diagram b from (7) is proportional to T^{-1} , and diverges as $T \rightarrow 0$. To the summation of such divergences (see Ref. 18) corresponds the series

in which each diagram contains a coefficient of the form $T^{-n}/n!$. The expression for the sum of this series has the form

$$P^{\gamma} = \frac{e^{-\beta\bar{\varepsilon}_f}}{1 + (2J+1)e^{-\beta\bar{\varepsilon}_f}}, \quad \bar{\varepsilon}_f = \varepsilon_f + 2J\rho g^2 \ln \frac{D}{|\varepsilon_f|},$$

i.e., corresponds to the replacement of ε_f by $\bar{\varepsilon}_f$ in the expression for P_0^{γ} of the diagram a . To similar results leads the removal of the divergences of the type T^{-1} in the Heisenberg model.^[15]

A more careful analysis of the diagram series for $\langle X^{\gamma\gamma} \rangle$ shows that a renormalization of the level does occur in the expression, $(i\omega_n - \bar{\varepsilon}_f)^{-1}$, for the continuous line, and that K_0^{γ} in the diagram c should be replaced by the average over the states of the complete Hamiltonian:

$$K^{\gamma} = 1 - \frac{2J}{2J+1} n_f.$$

As a result, in the limit $T \rightarrow 0$ we have for n_f the follow-

ing expression:

$$n_f = \theta(-\bar{\epsilon}_f) + \rho g^2 K(2J+1)/\bar{\epsilon}_f. \quad (10)$$

For the mean number of s electrons, we have from (8) the expression

$$n_s = \rho m \mu, \quad (11)$$

where m is the total number of degenerate electron bands. In the case of Ce there are 12 degenerate bands lying between the $5d^1$ and $6s^2$ atomic levels. We assumed that only six of them, with $J=5/2$, hybridize with the f level.

Finally, from (2), (3), (10), and (11) we obtain in the case of Ce the following expressions for $\Pi(0, 0)$ and the square of the velocity of sound:

$$\Pi(0, 0) = -6\rho(2 + Kg^2/\bar{\epsilon}_f^2), \quad (12)$$

$$C^2 = \frac{1}{M} \left[\frac{1}{6\rho} (Z - n_f)^2 \frac{\bar{\epsilon}_f^2}{2\bar{\epsilon}_f^2 + Kg^2} + \frac{2b}{\Omega_0} (Z - n_f) \right], \quad (13)$$

where M is the ion mass.

As can be seen, the role of hybridization in the computation of $\Pi(0, 0)$ led to the smooth dependence $n_f(\bar{\epsilon}_f)$, which makes a contribution to the density of states in a finite range of $\bar{\epsilon}_f$ values.

The change in $C(P)$ occurring in the transition in question may be connected with the changes under pressure in the quantities $Z - n_f$, ρ , $\bar{\epsilon}_f$, and Ω_0 . Experimentally, it is well known¹⁹ that in regions far from the transition (i.e., for $|\bar{\epsilon}_f| \gg g$) the value of the quantity C in the γ phase virtually does not differ from the value in the α phase. Therefore, it can be seen from (12) that the quantity

$$(Z - n_f)^2 / (2\rho + 2b(Z - n_f)/\Omega_0)$$

virtually does not change in the region of the transition.

Thus, it can be assumed that the change in $C^2(P)$ should be determined by the position of the level $\bar{\epsilon}_f$, which changes its sign during the phase transition. The experimentally observed asymmetry in the minimum is connected with the presence of K : a smoother decrease for $K=1/6$ in the γ phase and a steeper one when $K \approx 0.73$ in the α phase. The neglect of the correlation of the f electrons at the same site corresponds to the replacement of K by unity.

Comparing (13) with the experimental data given in Refs. 9 and 10, we find for the dimensionless parameter ρg the value $\rho g \sim 0.03 - 0.05$; this value agrees in order of magnitude with the value $\rho g = 0.02$ obtained in Ref. 19

on the basis of an analysis of the phase diagram. Analysis of the experimental curve for $C(P)$ given in Ref. 9 with the aid of the formula (13) yields the following values for $\bar{\epsilon}_f/g$: at the transition point on the side of the γ phase $\bar{\epsilon}_f/g = -0.5$, while on the side of the α phase $\bar{\epsilon}_f/g = 1.4$.

Thus, the analysis carried out above shows that the correct allowance for the hybridization and the correlation of the f electrons within the framework of a relatively simple model describes quite well the behavior of $C(P)$ and yields realistic values for the phase-transition parameters.

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