Raman scattering on superconductors in the presence of charge-density states

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Raman-active phonon modes close to twice the superconducting-gap energy yield a hybrid state, also Raman active, between the bare phonon and the superconducting-gap excitations. This result, which is obtained for finite temperatures, holds true if screening is taken into account.

Raman scattering experiments[1-3] on superconductors in the presence of low-frequency charge-density modes show evidence for a peak in the Raman spectrum near twice the gap energy. Only in the charge-density state phonon modes corresponding to wavelengths of 10–50 cm and a 2Δ of 10^{-4} to 10^{-3} eV exist. These important findings were explained by an interesting calculation by Balseiro and Falicov[4] (also compare Schuster[5] and Machida[6]), who considered the phonon state as a hybrid between the bare phonon and electron-hole pairs of superconducting-gap excitations. This calculation used standard perturbation techniques[7] for the evaluation of the phonon propagator, but did not take into account the Coulomb screening of the electron-phonon coupling (see Fig. 1).

Littlewood and Varma[8,9] and subsequently Browne and Levin[10] questioned these results because of nonscreening and argued that the effect disappears if screening is taken into account. A recent paper of Klein and Dierker[11] concentrates on phonon dispersion for nonvanishing momenta and, in the limit of $q \rightarrow 0$, seems to agree with Balseiro and Falicov. Because of the criticism mentioned above, we reopen the discussion to test Balseiro and Falicov's calculation by inclusion of charge screening. Furthermore, the lowest-order contribution to the phonon polarization in the superconducting state is enumerated for finite temperatures. As a result we find it still possible to see a resonance from the hybrid mode when Coulomb shielding is taken into account.

We begin with the calculation of the phonon Green's function defined by Dyson's equation[7]

$$D^{-1}(\omega) = D_0^{-1}(\omega) - \Pi(\omega) \ , \tag{1}$$

which yields the dispersion relation for the hybrid state at the frequency Ω by setting $D^{-1}(\Omega) = 0$. $\Pi(\omega)$ is the irreducible (proper) phonon polarization and contains the electron-phonon coupling g_0, which is screened by Coulomb interaction. It can be developed in a diagrammatical expansion (Fig. 2) and summed up to give

$$\Pi(\omega) = g_0^2 [1 - VP(\omega)]^{-1} P(\omega) \ . \tag{2}$$

In the lowest-order (random phase) approximation, $P(\omega)$ is the electron-hole loop of superconducting-gap excitations, which was first calculated by Schuster[5] for zero temperature. V is proportional to a photon propagator and is approximated by the Coulomb potential. Insertion of $D_0(\omega) = 2\omega_0 \times (\omega^2 - \omega_0^2 + i\delta)^{-1}$ yields the following implicit equation for the frequency Ω of the hybrid state:

$$\Omega = \omega_0^2 - [(\omega_0^2 - \Omega^2) V - 2g_0^2 \omega_0] P(\Omega) \ ,$$
$$P(\Omega) = -8N(0)\Delta^2 \int_0^\infty dx \frac{\coth[\beta\Delta\cosh(x)]}{4\Delta^2[\cosh(x)]^2 - \Omega^2} \ . \tag{3}$$

$N(0)$ is the density of the electron states at the Fermi surface, the gap $\Delta(T) = \Delta(T=0)(1 - T/T_c)^{1/2}$ is assumed to be independent of frequency and $\beta = (k_B T)^{-1}$. Equation (3) reduces to the result of Balseiro and Falicov if terms containing the Coulomb potential V are neglected.

At this point it is necessary to compare the relative strength of the Coulomb potential V and the electron-phonon potential $2g_0^2/\omega_0$. However, it is not possible to use the form $V(q) = 4\pi e^2/q^2$, since it diverges for small q^2. On the other hand, a dispersion relation $cq = \omega_0$ as for an acoustic phonon mode cannot be assumed. Nevertheless, the Eliashberg theory[7] provides a qualitative hint. It states

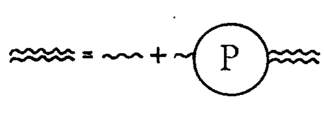

FIG. 1. Phonon propagator, as calculated by Balseiro and Falicov (Ref. 4).

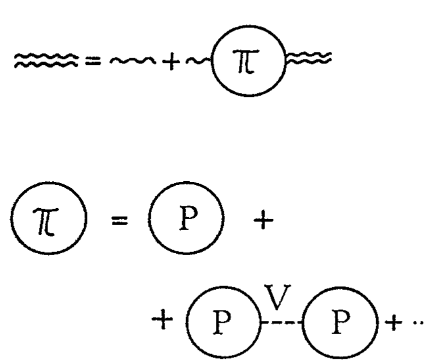

FIG. 2. Phonon propagator with Coulomb screening.

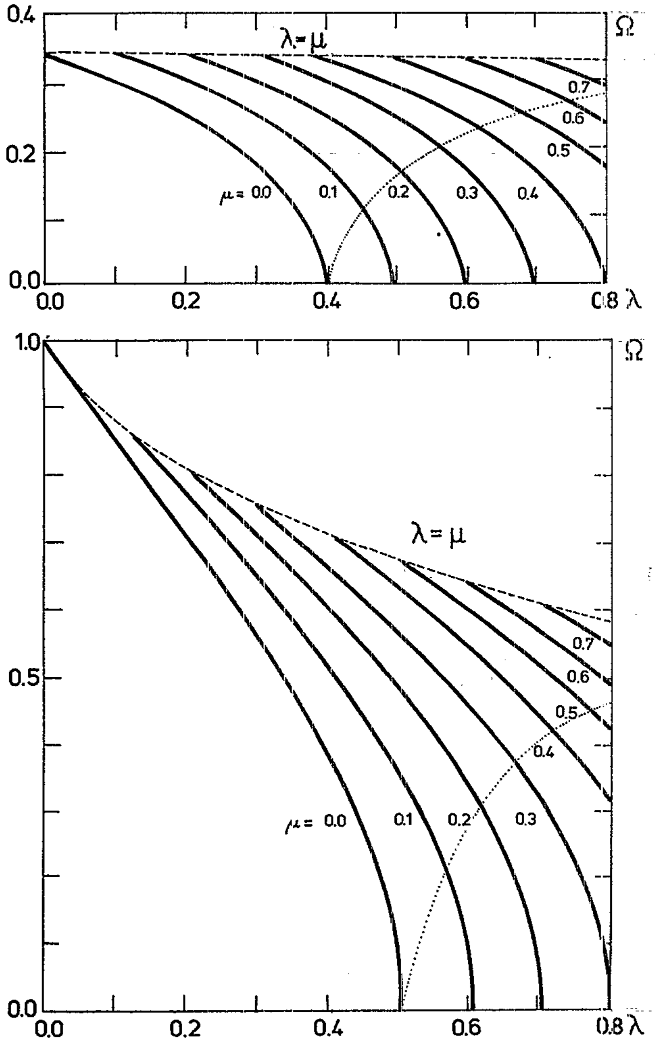

FIG. 3. Frequency of the hybrid state as a function of λ for fixed values of μ for $T=2$ K in units of ω_0. Dashed lines indicate values where $\lambda=\mu$. Dotted lines indicate values of λ and μ from the Eliashberg theory. (a) shows $\Delta=0.2\omega_0$ and (b) shows $\Delta=0.8\omega_0$.

that, for the material to be a superconductor, the average Coulomb coupling strength between electrons has to be less than the average phonon coupling strength. Therefore, the parameters $\mu=VN(0)$ and $\lambda=2g_0^2N(0)/\omega_0$ can be introduced as parameters in formal analogy to the Eliashberg theory. However, these parameters must not be considered identical with their averages, but as arbitrary coupling constants. In Fig. 3 the frequency of the hybrid state is plotted as a function of λ for different values of μ at $T=2$ K. For $\mu=0$ the results of Balseiro and Falicov are regained. The region below the dashed line is the Bardeen-Cooper-Schrieffer pairing condition $\lambda-\mu>0$ for every momentum and frequency rather than an average. The dotted line indicates Ω for allowed values of λ and μ obtained by McMillan's[12] equation for T_c, taking $T_c=7.2$ K and $\theta_D=210$ K for $NbSe_2$ from Refs. 1 and 13. Here, an Einstein spectrum with the frequency ω_0 had to be assumed, which is a too ideal assumption for the materials considered.

From the numerical work the following results are obtained.

(i) For small $\lambda-\mu$ (up to values of 0.2), Ω gets close to 2Δ, if $2\Delta\leq\omega_0$.

(ii) If $2\Delta>\omega_0$ and $\lambda-\mu$ is small as above, Ω tends to ω_0.

(iii) For larger values of $\lambda-\mu$ (from 0.3–0.5), Ω drops to zero.

(iv) Inclusion of Coulomb screening improves convergence of Ω towards 2Δ or ω_0.

In conclusion, the calculations show that although Littlewood and Varma's criticism of the explanation of Balseiro and Falicov is correct, the effect does not go away. The standard electron-phonon coupling can be applied to gain the observed peaks in Raman scattering experiments on superconductors when charge-density modes are present.

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