Comment on "Nonmonotonic $d_{x^2-y^2}$ Superconducting Order Parameter in Nd_{2-x}Ce_xCuO₄"

In a recent Letter [1], Blumberg *et al.* address the symmetry of the superconducting gap in cuprate superconductors. In particular, in the electron-doped systems the issue is not yet settled, and not even the phase sensitive experiments [2] arrive at a consistent conclusion. Therefore more spectroscopic information is useful: In addition to the B_{1g} and B_{2g} Raman spectra [3], Blumberg and co-authors measured the A_{1g} component by using an excitation energy of 1.9 eV. They obtain positions of the A_{1g} , B_{1g} , and B_{2g} pair breaking peaks at 45, 50 and 67 cm⁻¹, respectively, and conclude, from these positions alone, that the superconducting order parameter has $d_{x^2-y^2}$ symmetry with a nonmonotonic dependence on the azimuthal angle ϕ [see Fig. 1(b) of Ref. [1]].

In this Comment, we show that the basis for this conclusion is insufficient. This becomes already clear by just following the qualitative arguments of the authors: Since the Raman scattering amplitudes $\gamma_{\mu}(\phi)$ of all symmetries μ are finite at the maximum Δ_0 of the proposed gap function, the spectra in *all* symmetries will exhibit structures at the same energy $2\Delta_0$ as opposed to what is observed [1,3]. In addition, if $\Delta(\phi)$ has components up to $\sin(10\phi)$ (see caption of Fig. 1) as proposed in Ref. [1], it is hard to probe them with $\gamma_{\mu} \propto \sin(2\phi)$ without applying a model. Therefore, we calculated the Raman response

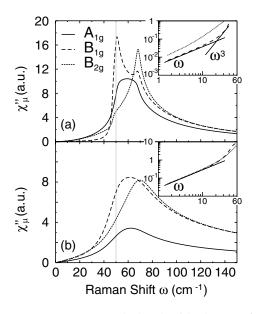


FIG. 1. Raman response calculated with the gap function proposed in Ref. [1]. The best analytical approximation is given by $\Delta(\phi) = \Delta_0[\sin(2\phi) + a_1\sin(6\phi) + a_2\sin(10\phi)]$ $(2\Delta_0 = 67 \text{ cm}^{-1}, a_1 = 0.42, a_2 = 0.17)$ using the same definition of ϕ as in Ref. [1]. In this nomenclature, the scattering amplitudes read $\gamma_{B_{1g}} \propto \sin(2\phi)$, $\gamma_{B_{2g}} \propto \cos(2\phi)$, and $\gamma_{A_{1g}} \propto -\cos(4\phi)$. Neither for small (a) nor for large (b) damping can the power laws, in particular, the ω^3 dependence in B_{1g} symmetry, be observed any more for $\omega < 20 \text{ cm}^{-1}$ (insets).

explicitly [4]. For a small phenomenological damping $\Gamma/\Delta_0 = 0.04$ [Fig. 1(a)], both the B_{1g} and the B_{2g} spectra exhibit two structures at around 50 and 67 cm⁻¹, and the A_{1g} peak is at approximately 60 and not at 45 cm⁻¹ as in the experiment. Even for this small damping, all spectra are linear [see inset of Fig. 1(a)] up to approximately 20 cm⁻¹ [4]. A larger damping $\Gamma/\Delta_0 = 0.30$ [Fig. 1(b)] not only smears out the double peak structure but also completely kills the power laws characteristic for *d*-wave pairing for this type of gap. In general, neither the **k** dependence nor the magnitude of the gap can be derived from the peak positions alone. Rather an appropriate model and the complete study of the low-temperature and low-frequency power laws are required. Then, constraints for the gap similar to those from the specific heat or the magnetic penetration depth could be obtained.

Here, it is indeed the magnetic penetration depth [5] $\lambda(T)$ which is in direct conflict with the proposed form of the gap. The temperature dependence of $\lambda(T)$ can be readily calculated from the functional dependence $\Delta(\phi)$. No agreement with the data [5] at any doping can be achieved. Supposing the nonmonotonic gap would be realized an analysis with a monotonic one [5] would lead to $\Delta_0 \simeq 9k_BT_c$ in spite of the restricted phase space around the node.

In conclusion, the functional dependence of the gap proposed in Ref. [1] is neither sufficiently supported by the Raman results nor compatible with the magnetic penetration depth. In spite of that, an anisotropic s-wave as suggested earlier [3] is probably not the full story either, at least not in the entire doping range. Therefore the issue of the superconducting gap in the electron-doped systems cannot at all be considered solved by now.

- F. Venturini and R. Hackl Walther Meissner Institute Bavarian Academy of Sciences 85748 Garching, Germany
- U. Michelucci EKM Universität Augsburg 86135 Augsburg, Germany

Received 6 May 2002; published 7 April 2003 DOI: 10.1103/PhysRevLett.90.149701 PACS numbers: 74.25.Gz, 74.72.Jt, 78.30.-j

- [1] G. Blumberg et al., Phys. Rev. Lett. 88, 107002 (2002).
- [2] L. Alff *et al.*, Phys. Rev. Lett. **83**, 2644 (1999); C. C. Tsuei and J. R. Kirtley, Phys. Rev. Lett. **85**, 182 (2000).
- [3] B. Stadlober et al., Phys. Rev. Lett. 74, 4911 (1995).
- [4] T. P. Devereaux and D. Einzel, Phys. Rev. B 51, 16336 (1995); T. P. Devereaux, Phys. Rev. Lett. 74, 4313 (1995);
 F. Venturini *et al.*, Phys. Rev. B 62, 15204 (2000).
- [5] J. D. Kokales *et al.*, Phys. Rev. Lett. **85**, 3696 (2000); J. A. Skinta *et al.*, Phys. Rev. Lett. **88**, 207003 (2002); **88**, 207005 (2002).