Inelastic Incoherent Neutron Scattering as an Alternative Method to Investigate Local Critical Behavior

T. Schneider and P. F. Meier
IBM Zurich Research Laboratory, 8803 Rüschlikon, Switzerland
(Received 5 June 1973)

In this paper we establish the relation between the spectral density of the spatial autocorrelation function as probed by electron-spin resonance and the inelastic incoherent differential neutron scattering cross section. Special emphasis is placed on the critical phenomena associated with structural phase transitions.

In recent years the critical dynamics of structural phase transitions has been investigated quite extensively by means of neutron scattering[1-3] and electron-spin resonance,[4,5] and more recently using light scattering.[6,7] An interesting observation is the discovery of a central peak in addition to the conventional soft-mode spectrum in the dynamic form factor. The physical origin of this central peak is, however, still a matter of controversy.[8]

Interpretation of neutron scattering experiments has so far been restricted to a discussion of

$$S_{\alpha\alpha}(\vec{k},\omega)=\int_{-\infty}^{+\infty} dt\, e^{-i\omega t}\,\langle\varphi^{\alpha}(\vec{k},t)\,\varphi^{\alpha}(\vec{k},0)\rangle \qquad (1)$$

where, in the case of $SrTiO_3$, $KMnF_3$, and $LaAlO_3$ at $T \geq T_c$,

$$\varphi^{\alpha}(k,t)=\frac{1}{N}\sum_{l} e^{i\vec{k}\cdot\vec{R}^{l}}\,\varphi_{l}^{\alpha}(t) \;. \qquad (2)$$

φ_l^α corresponds to the rotation angle of the BO_6 octahedra in the lth unit cell around direction α. $S(\vec{k},\omega)$ is proportional to the coherent-scattering cross section.[9] The incoherent-scattering contribution has been treated as giving rise to a smooth and uncritical background.[1-3,8]

The electron-spin-resonance method, on the other hand, probes locally in space.[4,5] As a consequence, information is obtained on the spectral density of the spatial autocorrelation function, namely,

$$\sum_{k} S_{\alpha\alpha}(\vec{k},\omega)=\int_{-\infty}^{+\infty} dt\; e^{-i\omega t}\,\langle\varphi_{l}^{\alpha}(t)\;\varphi_{l}^{\alpha}(0)\rangle \;. \qquad (3)$$

An important point of this note is to recall that $\sum_k S(\vec{k},\omega)$ is proportional to the incoherent neutron scattering cross section. In fact, the total differential cross section is given by[9]

$$\frac{d^2\sigma}{d\Omega\, d\omega}\sim\langle b\rangle^2\; S(\vec{k},\omega)+(\langle b^2\rangle-\langle b\rangle^2)\,\frac{1}{N}\sum_{k} S(\vec{k},\omega). \qquad (4)$$

b is the scattering length of the nuclei, associated with the particular rotation. The first and second terms correspond to coherent and incoherent inelastic scattering, respectively. We note that the electron-spin-resonance data support the following form of $\sum_k S(\vec{k},\omega)$ close to T_c,[4,5]

$$\sum_{k} S_{\alpha\alpha}(\vec{k},\omega)=S_{\alpha\alpha}^{ll}(\omega)=\frac{S_{\alpha\alpha}^{ll}(t=0)}{\pi}\;\frac{\omega_R}{\omega^2+\omega_R^2}\;, \qquad (5)$$

where

$$S_{\alpha\alpha}^{ll}(t=0)=\langle(\varphi_{\alpha}^{l})^2\rangle=\int_{-\infty}^{+\infty} d\omega\sum_{\vec{k}} S_{\alpha\alpha}(\vec{k},\omega)\;. \qquad (6)$$

ω_R is a relaxation frequency. Theoretical investigations predicting that ω_R tends to zero by approaching T_c[10,11] are supported by recent experiments.[4,5] In view of this, the incoherent-scattering contribution is expected to become critical near $\omega = 0$. In fact, from Eq. (5) we find

$$\sum_{k} S_{\alpha\alpha}(\vec{k},0)=\frac{\langle(\varphi_{\alpha}^{l})^2\rangle}{\pi}\;\frac{1}{\omega_R}\;. \qquad (7)$$

ω_R tends to zero by approaching T_c, whereas $\langle(\varphi_\alpha^l)^2\rangle$ remains finite and nonzero and is equal to $(0.4^\circ)^2$ in $SrTiO_3$.[4,5]

This analysis reveals:

(i) Incoherent neutron scattering is an alternative method to investigate the local dynamic behavior near structural phase transitions, provided that $\langle b^2\rangle-\langle b\rangle^2\neq 0$.

(ii) The differential incoherent-scattering cross section is expected to become critical. Consequently, the assumption of a smooth and regular inelastic and incoherent background contribution is no longer guaranteed.

(iii) The differential incoherent cross section does not depend on the momentum transfer [Eq. (5)]. Due to the variables chosen, however, our equations are applicable only for those values of $\vec{k}$ where $\varphi^{\alpha}(\vec{k})$ represents a rotation.

Finally, we note that the incoherent-scattering cross section may be increased upon doping with suitable isotopes.

We acknowledge stimulating discussions with J. D. Axe, M. Blume, P. Heller, K. A. Müller, S. M. Shapiro, and E. Stoll.

[1]T. Riste, E. J. Samuelson, K. Otnes, and J. Feder, Solid State Commun. **9**, 1455 (1971).
[2]G. Shirane and J. D. Axe, Phys. Rev. Lett. **27**, 1803 (1971).
[3]S. M. Shapiro, J. D. Axe, G. Shirane, and T. Riste, Phys. Rev. B **6**, 4332 (1972).
[4]Th. von Waldkirch, K. A. Müller, and W. Berlinger, Phys. Rev. B **7**, 1052 (1973).
[5]K. A. Müller, in Proceedings of the NATO Advanced Study Institute on Anharmonic Lattices, Structural Transitions and Melting, Noordhoff, 1973 (unpublished).
[6]E. F. Steigmeier and N. Auderset, Solid State Commun. **12**, 565 (1973).
[7]E. F. Steigmeier, H. Auderset, and G. Harbeke, Solid State Commun. 12, 1077 (1973).
[8]See, for example, Ref. 5.
[9]W. Marshall and S. W. Lovesey, *Theory of Thermal Neutron Scattering* (Clarendon, Oxford, England, 1971).
[10]F. Schwabl, Phys. Rev. Lett. **28**, 500 (1972); Z. Phys. **254**, 57 (1972).
[11]T. Schneider, Phys. Rev. B **7**, 201 (1973).