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2004 J. Phys.: Condens. Matter 16 3869

(http://iopscience.iop.org/0953-8984/16/23/007)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **16** (2004) 3869–3878 PII: S0953-8984(04)74052-7

Anomalous displacement of magnetic Bragg peaks in the presence of short range order

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Received 19 December 2003, in final form 18 February 2004
Published 28 May 2004
Online at stacks.iop.org/JPhysCM/16/3869
DOI: 10.1088/0953-8984/16/23/007

Abstract

An asymmetric shift in the position of the magnetic Bragg peak with respect to the fiducial lattice has been observed by resonant x-ray scattering in a diverse series of antiferromagnetic compounds. A possible explanation is given in terms of a generalized Berry phase correction.

Since the pioneering work of Shull and collaborators [1] neutron diffraction has been the probe of choice for observing magnetic diffraction peaks and examining the evolution of the magnetic sub-lattice order on the atomic scale. When the long range order (LRO) in the AF state is of a different periodicity from that of the underlying chemical lattice, peaks arise at new, previously forbidden, positions in the diffraction pattern. As a result, in order to preserve Bragg's Law, it is necessary to introduce an explicit phase variable, $\phi = \mathbf{Q}_0 \cdot \mathbf{r}$, into the defining magnetic order parameter where $\mathbf{Q}_0$ is the antiferromagnetic wavevector.

In this paper, we detail studies of the magnetic diffraction peaks recorded in a number of materials which, by neutron diffraction, are known to condense into a state of antiferromagnetic (AF) order below the Néel temperature, T_N. As a complement to the information accrued by neutron diffraction, in the following we exploit the potentials of resonant x-ray magnetic scattering. In particular, the high wavevector resolution and incident flux combined with

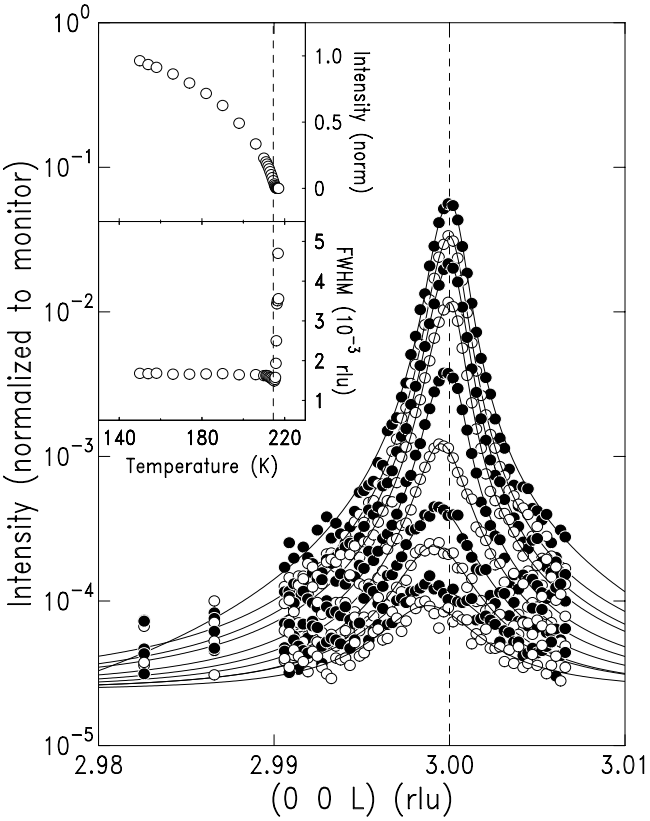

Figure 1. Diffraction data from the (003) AF reflection of USb in the vicinity of T_N at the U M_4 edge (3.728 keV at XMaS, ESRF). Top scan at $T = 214.5$ K, next at 215.0 K, then subsequent scans at intervals of 0.25 K. The lines are Lorentzian squared functions to fit the data; note the logarithmic scale of intensity. (Any smooth function fitted to the data may be used to quantify the shift and width as a function of temperature.) Top inset—the integrated intensity of the AF (003) as a function of temperature. Lower inset—the full-width at half maximum (fwhm) as a function of temperature.

polarization analysis available at the European Synchrotron Radiation Source, Grenoble (ESRF), have enabled us to observe a change in effective magnetic periodicity near T_N as measured by an asymmetric shift in the Bragg wavevector *relative* to that of the underlying chemical lattice in a set of thermodynamically diverse materials. Such an effect was, to our knowledge, first noted in an earlier study of UO_2 by resonant x-ray scattering at the uranium M_4 energy [2], and at about the same time in HoB_2Ni_2C using photons tuned to the Ho L_3 edge [3]. To date no explanation has been advanced for these observations.

We illustrate this effect with data, figure 1, taken by resonant x-ray scattering at the U M_4 edge on the XMaS beamline at the ESRF, on a single crystal of USb. The small, but significant, shift in wavevector, recorded here at the magnetic (003) specular reflection, lies comfortably outside the resolution width of the spectrometer, and shows a characteristic drift to lower values with increasing temperature. USb, in contrast with UO_2 which undergoes a discontinuous change of magnetic phase at 30 K [2], exhibits a continuous transition [4] as demonstrated by the temperature dependence of the antiferromagnetic scattered intensity in the upper inset and the evolution of the magnetic q-widths plotted in the lower inset of figure 1.

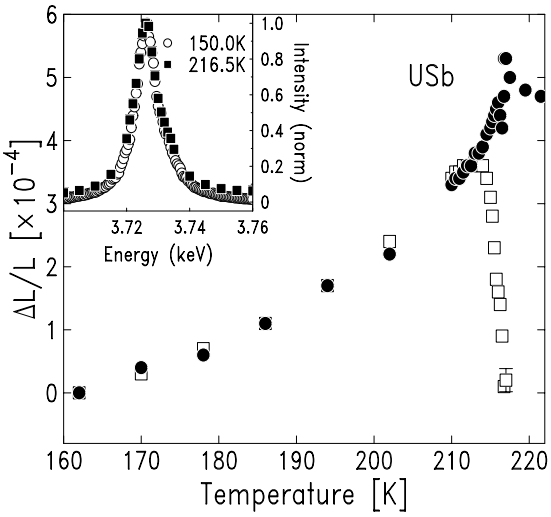

Figure 2. Wavevector shifts (divided by the low temperature L index) of charge (002) (filled points) and magnetic (003) (open points) for USb as a function of temperature (error bars $\sim$ size of points). Inset—normalized magnetic intensity as a function of incident photon energy for 150 K (open) and 216.5 K (solid points). The profiles are identical within experimental error, fwhm $\sim$ 9 eV, despite an intensity ratio of some three orders of magnitude, indicating that they originate from a common bulk volume [5] thereby eliminating the possibility that the observed shift, $D\tilde{Q}$, is primarily due to near surface effects.

The normalized shift in wavevector is plotted as a function of temperature in figure 2 together with that for the *charge* (002) reflection measured under the same experimental conditions, i.e. sampling the same absorption and a similar scattering volume. As the figure shows, the magnetic and charge peaks are commensurate throughout the ordered region (i.e. the two peak positions scale) despite the large magnetostrictive effects. However, as noted in figure 1, in the vicinity of T_N there is a *decrease* in the observed magnetic wavevector occurring in *opposition* to the lattice contraction, which continues unabated to a small anomaly around 216 K. In contrast with the magnetic reflection, no observable changes occur in either the intensity or width of the charge (002) reflection on passing through T_N, consistent with the magnetic structure being a 3**k** arrangement [4] having no crystallographic distortion from cubic symmetry below T_N.

Amongst the series of materials investigated, similar effects are seen, for example, in the heavy fermion superconductors UPd_2Al_3 and URu_2Si_2. Here, as illustrated in figure 3 for URu_2Si_2, the shifts in the magnetic wavevector lie outside the narrow charge peak profiles. This makes it difficult to propose the idea that a small strained (i.e. with different lattice parameter) scattering volume near the surface exhibits magnetism. Such scenarios, associated with possible second-length scales [5, 6], also have the difficulty that the charge and magnetic peaks in all cases have been measured with photons of the same energy. As noted, the scattering volumes are thus similar and it is the position of the magnetic *with respect to* the charge peaks that shows an anomalous shift as is vividly demonstrated in USb. Furthermore, any theory invoking strain has to account for the change in the wavevector over a narrow temperature range.

The inset to figure 2 shows the magnetic intensity as a function of incident photon energy well below and in the neighbourhood of T_N for USb. Similarly, data are given in the inset of figure 3 for URu_2Si_2. If the observed shift in magnetic wavevector were driven by either near surface effects, or by changes in the density of polarized states near T_N, the width and profile

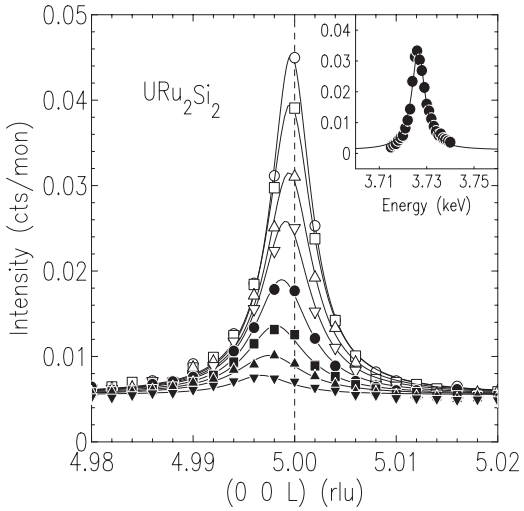

Figure 3. Longitudinal scans through the (005) magnetic specular reflection in URu_2Si_2 showing the shift and broadening of the peak. The highest intensity is with $T = 10$ K, and other profiles correspond to temperatures as in figure 4 below. Inset—incident photon energy dependence of the (005) peak at $T = 10$ K. The wide Lorentzian profile is independent of temperature and signifies a substantial intermediate state matrix element associated with a strong $5f$ polarization whilst the broad q-vector peak (main frame) gives a measure of the short-range magnetic order.

of the energy dependences would be modified [5]. No changes are seen. Thus the energy resonances, the independence of effects to the surface preparation and order of the transition all argue against near surface sensitivity as the underlying cause.

In figure 4 we show the change of the magnetic wavevector, $D\tilde{Q}$, as a function of temperature for UO_2 (top frame), UPd_2Al_3 (middle frame) and URu_2Si_2 (lower frame) together with that of the lattice parameter. Independent of being metal or insulator, the continuous or discontinuous nature of the magnetic transition and the magnitude of T_N the similarities across this heterogeneous set of materials are striking. Thus material specific details such as magnetic phase heterogeneity near T_N at a discontinuous transition, dielectric or magnetic screening, or the presence of lattice vibrations are unlikely to be fundamental to understanding this phenomenon. These conclusions are supported by parity considerations, which eliminate both the build up of (energy integrated inelastic) thermal diffuse scattering and incommensurate ordering which would give a pair of symmetrically displaced peaks around each reflection.

Summarizing the experimental situation, two important observations appear generic to the anomalous shift: (a) $D\tilde{Q}$ is in each case associated with a broadening of the magnetic diffraction peak and (b) the shift is to smaller wavevector. Independent of details, the crucial wavevector shift in magnetic scattering of a given volume *relative* to the charge appears to lie outside interpretation within conventional diffraction theory.

We commence our appraisal with a brief recapitulation of the conditions necessary for coherent diffraction. The essential observation is that lattice periodicity gives rise to momentum quantization, and hence to discrete Bragg peaks in momentum space. This focus brings out the fundamental role of the system's periodic Hamiltonian and boundary conditions in the formulation of the scattering amplitude from a periodic array. The spatial symmetry both defines the acceptable state vectors underlying the appropriate Hilbert space and the class of operators permitted in this space. On scattering, the probe and crystal enter into a joint state vector, $|\Psi\rangle$, which is most frequently represented by fragmentation into non-interacting

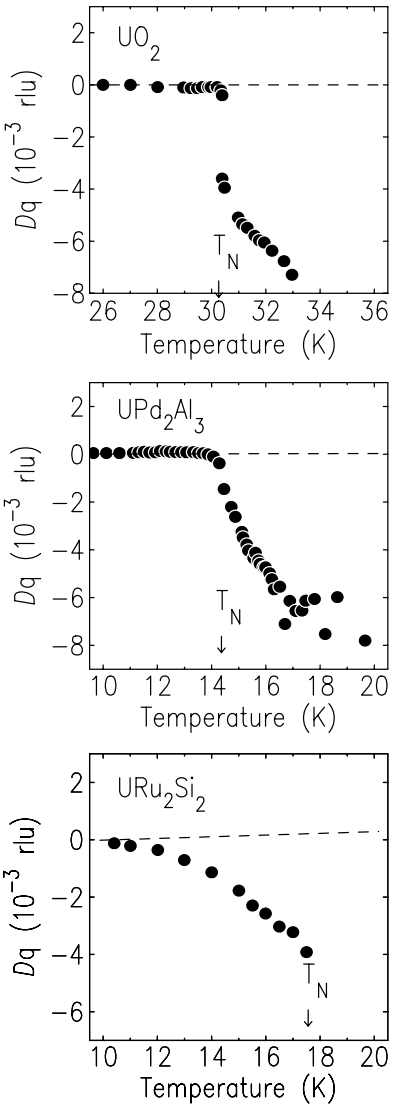

Figure 4. Shift, $D\tilde{Q}$, in the reciprocal lattice position of the *magnetic* Bragg peak recorded at the (110) reflection in UO_2 [2], the (0 0 1.5) reflection in UPd_2Al_3 (both taken at X22C, NSLS, BNL) and the (005) reflection in URu_2Si_2 (taken at ID20, ESRF), all using photons tuned to the uranium M_4 edge at 3.728 keV. The change in the lattice parameter, as measured in the same experiment at the *charge* Bragg reflections, is indicated by the broken lines. In UO_2 the phase transition is known to be discontinuous [2].

primitive functions of state for the crystal, $|\phi\rangle$, and probe, $|\chi\rangle$, respectively together with a mandatory interaction (as seen through the cross section and also expressed in the primitive state vector weighing coefficients) introduced to resurrect at least some of the amplitude–phase correlations present in $|\Psi\rangle$. The decomposition may be represented by an entangled state of generic form, $|\Psi\rangle \approx \sum_i |\phi_i\rangle|\chi_i\rangle$, where the sum extends over all Bragg points compatible with the experimental situation. Bragg's insight on coherent diffraction phenomena, which predated the quantum picture, may be restated as a condition of stationary phase on the joint sub-systems.

An experimentalist manipulates the incident beam and records counts in a detector, i.e. has access only to the scattered part of the state vector; thus one is obliged to integrate out the crystal co-ordinates and treat the crystal as 'part of the universe'. Such a situation, under the Bragg constraint on acceptable state vectors, suggests that information maybe held in the *phase* of the scattered state vectors. Berry's influential paper of 1984 [7] highlighted the role of such symmetry constraints in creating a class of observables, given by the gauge invariant phase of the quantum mechanical amplitude, in addition to those given by the weight of its expectation value. The initial arguments of Berry have been extended over the years in many directions [8], of import to us here is the recognition that a closed path in parameter space may be realized not only by an explicitly cyclic variation of a parametric Hamiltonian, as considered for example in the Aharonov–Bohm effect [9] or in the (dynamic) Jahn–Teller distortion of polyatomic molecules [10], but also by a physically enforced symmetry as in a coherent scattering event.

Early investigations on the spatial symmetry constraints of an infinite periodic lattice were made by Zak and others [11, 12] introducing the concept of a non-cyclic, 'open path' geometric phase. In the following we investigate the role that the enforced symmetry of the coherent scattering event may have on diffraction phenomena. A generalized co-ordinate, β, is used to parameterize the evolution of the scattering event. As β goes from $\beta_{\rm i} \rightarrow \beta_{\rm f}$, the basic premise of coherent scattering is that the system state vector returns on itself, preserving phase information. This critical assumption, which appears to be borne out by the analysis given and its experimental verification (figure 5), may be cast as an open path geometric phase[10]. It is clear that the scattering interaction between probe and sample will modify both the phase and relative weight of the state vectors. Whilst the renormalization is a second order perturbative effect in the relative weighting coefficient, it appears as a first order correction to the phase. In the absence of symmetry constraints, the first order (Berry) phase term, is often justly ignored as it may be in situations where it amounts to a zero phase increment (modulo 2π). However, under particular experimental conditions, it may present itself as a non-trivial modification.

Coherent diffraction is characterized by the conditions of (i) a stationary phase of the scattering amplitude (since it is only in this case that contributions from different space–time volumes add constructively) and (ii) that the system should be found in its initial state after the event. The phase of the scattering process may be written as $\langle\psi_{\rm f}|H|\psi_{\rm i}\rangle = a{\rm e}^{-{\rm i}\Delta\varphi}$ where i, f refer to the initial and final states, respectively, H is the interaction Hamiltonian and $\Delta\varphi = [\Delta k_{\rm p} - q_{\rm l}]aN - [\Delta E_{\rm p} - \omega_{\rm l}]t - [\gamma_{\rm p} - \gamma_{\rm l}]$ is the total phase increment accumulated in $|\Psi\rangle$ over the coherent scattering space–time volume expressed in terms of the probe, (p), and lattice, (l), contributions. The lattice period is a, N the number of correlated lattice cells within the probe coherence volume along the direction $q_{\rm l}$ and t the coherence time of the scattering. The stationary condition sets $\Delta\varphi = 0$. Within Bragg's semi-classical model the invariance of the first two terms corresponds to the assumption of momentum and energy conservation, respectively. The last term sums to zero since an isolated system cannot have any overall phase dependency, i.e. the Berry phase increment of crystal is equal and opposite to that of probe. The coherent return to the initial state may be expressed as a function of the generalized co-ordinate β. The scattering system goes from an initial state $|\phi(\beta_i)\rangle$ to a final state $|\phi(\beta_f)\rangle$ with the condition $|\phi(\beta_f)\rangle = S^{-1}|\phi(\beta_i)\rangle$ where S expresses the invariance, implicit in a coherent scattering event between initial and final states in the crystal, i.e. $H_{\rm l}(\beta_f) = S^{-1}H_{\rm l}(\beta_i)S$.

For an infinitesimal change in scattering parameter the incremental change in $|\phi\rangle$ is calculated in perturbation theory as the weighted linear superposition of intermediate states.

[10] The scattering event is a one-off rather than cyclic event; for this reason the 'open path' terminology is used. Despite the common terminology, this article is not concerned with problem of a position variable in an infinite lattice which is addressed in [11, 12].

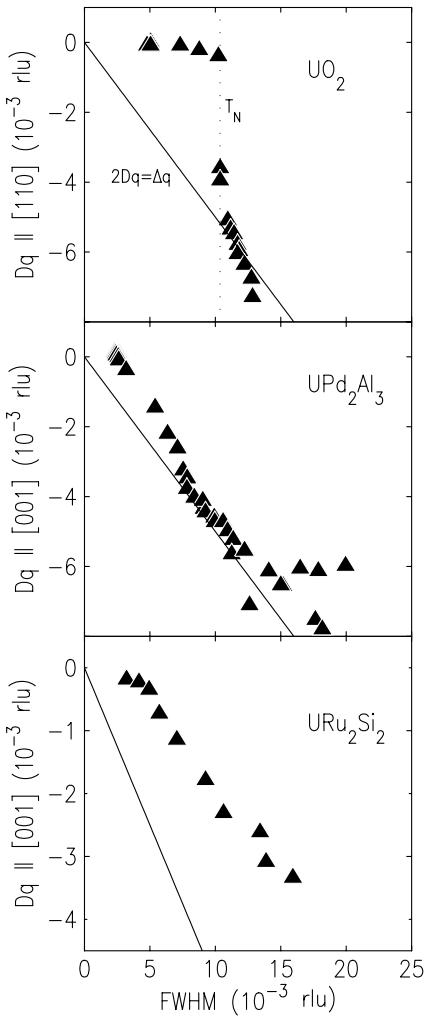

Figure 5. The anomalous displacement, $D\tilde{Q}$, of the antiferromagnetic wavevector as a function of fwhm in URu_2Si_2, UPd_2Al_3 and UO_2, with temperature as an implicit parameter. The solid lines of slope $-1/2$ are the predicted dependency of $D\tilde{Q}$ on the fwhm. Note that for URu_2Si_2, in the absence of LRO, the assignment of $D\tilde{Q} = 0$ at the base temperature of 10 K is arbitrary.

An implicit choice of gauge is made, corresponding with the perturbative additions being orthogonal to the initial state,

$$|\phi(\beta + \Delta\beta)\rangle \approx |\phi(\beta)\rangle + \sum_{\nu \neq \phi} |\nu(\beta)\rangle \frac{\langle \nu | H' | \phi \rangle}{E_\nu - E_\phi}\bigg|_\beta \Delta\beta$$

where $|\phi\rangle$ and $|\nu\rangle$ are, respectively, the initial and intermediate crystal state vectors, H' is the β derivative of H and explicit corrections involving normalization of states are temporarily neglected[11]. This parallel transport gauge, which leads to a local rotation of $|\phi\rangle$ as a function

[11] The weighting coefficient correction to the virtual state is of second order whilst its phase rotation is of first order in $\Delta\beta$. On assumption that the infinitesimal phase correction is of similar magnitude in excited and initial states, the major effects are taken care of by the global phase factor, $\exp(-i\Delta\gamma)$.

of β, is incompatible with the global phase invariance over β demanded by the condition of coherence and must be generalized [12]:

$$|\phi(\beta+\Delta\beta)\rangle \approx \mathrm{e}^{-\mathrm{i}\Delta\gamma}\left[|\phi(\beta)\rangle + \sum_{\nu\neq\phi}|\nu(\beta)\rangle\frac{\langle\nu|H'|\phi\rangle}{E_\nu - E_\phi}\bigg|_\beta \Delta\beta\right].$$

The phasing correction, which is applied uniformly to both the initial and virtual states, may be identified as an infinitesimal Berry phase increment, $\Delta\gamma = \mathrm{i}\langle\phi|\partial\phi/\partial\beta\rangle\Delta\beta$. In this way the rotation of $|\phi\rangle$ is corrected at each increment of β and, on completion with $\beta = \beta_f$, the state vector returns to its initial condition. The total corrective phase increment, summed over the space–time volume of the scattering event, γ_I, is passed to the probe through the condition of stationary phase and appears, for the coherent elastic scattering process, as a momentum shift in Bragg peak resonance condition.

The gauge corrected state vector which emerges is manifestly not normalized. However, the change in weighting coefficient appears in second order, $\approx\sqrt{1-\sum_{\nu\neq\phi}|\langle\nu|H'|\phi\rangle/(E_\nu - E_\phi)|^2(\Delta\beta)^2}$, and hence is neglected at each infinitesimal whilst the phase shift, $\Delta\gamma$, which arises from the local state vector rotation occurring in the second order of perturbation theory, manifests as a primary correction $\sim\Delta\beta$.

The distinction between the canonical phase wavevector $\tilde{Q}$ and kinematic wavevector Q_{mv} must be maintained. $\tilde{Q}$ is a property of the wavefunction, whilst Q_{mv} characterizes the kinematic nature of the state. For a free particle Q_{mv} is identified with the kinetic momentum and may be experimentally defined, for example, by the (macroscopic) geometrically determined space–time flight path of the probe. Thus for the probe, where there is only a contribution from the kinematic momentum, consistency demands,

$$\tilde{Q}_\mathrm{probe} = Q_{mv}.$$

For a periodic array Q_{mv} is identified with the mechanical lattice periodicity, i.e. the lattice or crystal momentum, which gives rise to discrete Fourier components (quantization of momentum in units of the reciprocal lattice vector) in coherent elastic scattering and yields,

$$\tilde{Q}_\mathrm{sample} = Q_\mathrm{lattice}.$$

In the quantum approach a condition of stationary phase is set, $\tilde{Q}_\mathrm{probe} = \tilde{Q}_\mathrm{sample}$, which finds its equivalent in the conservation of kinematic momentum, $Q_{mv} = Q_\mathrm{lattice}$. In this light, given the constraints of periodicity on the lattice functions of state, a primary role of a gauge invariant phase in the scattering process is not unexpected. In the presence of a Berry phase vector $\tilde{Q} = Q_\mathrm{lattice} - \partial\gamma/\partial R$ and it remains to fix the relative sign of the Berry phase vector, $D\tilde{Q} \equiv \partial\gamma/\partial R$, with respect to Q_lattice. We argue by analogy with a charged particle in a magnetic field: as the scattering interaction proceeds, and the impulse of momentum given by the probe propagates through the lattice, the system makes a best choice of wavefunction to *minimize* the phase momentum increment over the scattering path. Thus, to maintain a periodic gauge, the state vector appears to retard its phase by the corrective increment $\Delta\gamma = \mathrm{i}\langle\phi|\partial\phi/\partial\beta\rangle\Delta\beta$ at each step. This is consistent with the observation of a negative gradient in all graphs of figure 5.[12]

Spatially and temporally there are bounds on the largest unit capable of maintaining a coherent transition. Scattering, and with it phase accumulation, being arrested when the integrated phase adjustment, γ, approaches $\sim\pi$ at which point the coherence condition necessary for diffraction is lost. For resonant x-ray scattering the time of coherence is to be

[12] The parallel with Lenz's law has been noted and a counter example has yet to be found; however, it is not known if the relative opposition of phase is based in a general minimization principle.

superior to the resonant lifetime of the intermediate state and spatially it is taken to comprise N repeat units leading to the condition, $D\tilde{Q} \sim \pi/Na$. At the same time, spatial limitation of the coherent diffracting unit also breaks strict periodicity. This blurs the lattice momentum exchange giving a contribution to the intrinsic width of the diffraction peak, $\Delta q \sim 2\pi/Na$, from which one obtains the testable prediction, $D\tilde{Q} \approx (\gamma/2\pi)\Delta q$. Comparison with the experimental data is given in figure 5, where Δq is the measured full width half maximum of the Bragg peak. As is seen, for UO_2, UPd_2Al_3 and URu_2Si_2 there is satisfactory agreement with the predicted phase shift of $\gamma \sim \pi$, giving a slope of $-1/2$. However, whilst the slope for the weak antiferromagnetic–superconductor URu_2Si_2 is close to $-1/2$, it is evident that quantitative agreement is lacking. In this compound, where the wavevector shift exists at all measured temperatures *below* T_N, the setting of $D\tilde{Q} = 0$ in figures 4 and 5 is evidently arbitrary. An offset of $\sim -2 \times 10^{-3}$ rlu is suggested by the experimental plot of figure 5 which would, by smooth extrapolation in figure 4, suggest URu_2Si_2 to remain in a dynamic state down to significantly lower temperature. This inference may be of importance both in understanding the anomalous low magnetic moment inferred from neutron diffraction and the formation of the simultaneous antiferromagnetic–superconducting ground state [13].

In summary, we have presented data which evidences an asymmetric shift in magnetic scattering wavevector from a broad series of compounds including metals and insulators, which exhibit both continuous and discontinuous transitions. The unique aspect of these experiments is to have the lattice serving as an *in situ* fiducial marker of essentially infinite periodicity, i.e. $\tilde{Q}_{\text{Thompson}} = Q_{\text{lattice site}}$ enabling a differential measurement of the shift in the magnetic response arising from the magnetic correlations of finite length scale having $\tilde{Q}_{\text{magnetic}} = Q_0 - D\tilde{Q}$. This difference is perhaps most vivid in the example of USb, figure 2. The shifts are in all instances small but lie well beyond the resolution limits of modern x-ray diffractometers. Experiments are currently limited on account of intensity. Presumably, with improved sources, other edges can be used; in particular the rare earths may give many opportunities for experiments.

The understanding of the observed shifts in wavevector appears to require the consistent treatment of terms formally beyond the first order perturbation theory of diffraction as commonly employed. The current approach emphasizes the fundamental aspect of a stationary phase in coherent Bragg scattering and points to an incompatibility of the symmetry requirements of coherence with the parallel transport gauge of standard perturbation theory. Thus Berry's insight, that apparently second order renormalization effects may occur as a primary phase correction in an experiment accessing only an isolated part of an extended Hilbert space within which the vectors and operators are constrained by symmetry, appears to be fundamental. In the quantum picture the Berry shift γ appears as a corrective element in the condition of stationary phase, translated into Bragg's wave picture it corresponds to the shift $Q_0 \to Q_0 - D\tilde{Q}$ with $D\tilde{Q} = \gamma/Na$.

Acknowledgment

One of us, NB, wishes to thank Jozef Spalek for stimulating discussions on this topic.

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